#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of Date: January 13, 2000

Applicants: Bednorz et al. Docket: YO987-074BY

Serial No.: 08/303,561 Group Art Unit: 1751

Filed: 09/09/94 Examiner: M. Kopec

For: New Superconductive Compounds Having High Transition

Temperatures, Methods For Their Use and Preparation

Assistant Commissioner for Patents

Nashington, D. C. 20231

# SUBSTITUTE BRIEF

PPEAL TO THE BOARD OF APPEALS AND INTERFERENCE'S

Applicants hereby appeal, Pursuant to 37 C.F.R. 1.192(c), the Examiner's final rejection of Claims as set forth in the: RECEIVED
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Five Advisory Actions dated February 25, 1999; and Final Office Action dated June 25, 1998

### **REAL PARTY IN INTEREST**

The above-identified patent application has been assigned to the International Business Machines Corporation. The assignment has been recorded in the U.S. Patent and Trademark Office: Recordation date January 9, 1995; Reel/Frame 7331/0519.

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## **RELATED APPEALS AND INTERFERENCES**

There are no related appeals and interferences.

Co-pending US application serial number 08/479,810 filed on June 7, 1995 is a division of the present application.

### STATUS OF CLAIMS

Claims 24-26, 86-90 and 96-177 are presently pending in the present application.

Claim 136 is allowed. Claims 24-26, 86-90, 96-135 and 137-177 have been finally rejected in the Office Action dated June 25, 1998 which provides the basis for this appeal.

The following table lists the independent claims and the claims which depend therefrom:

Set	Independent Claim	Dependent Claim
1	24	25, 26
2	86	87
3	88	90
4	96	97-102
5	103	104-108
6	109	
7	110	
8	111	
9	112	
10	113	
11	114	
12 12	115	
11 12 13 14 15 16 17	116	
14	117	
15	118	
16	119	
17	120	
18	121	
19	122	
20	123	
21	124	770
22	125	
23	126	164 R 23 D 165 00 D 166 M
24	127	7 10
25	128	7 A
26	129	164 23 m
27	130	165 8 8 0
28	131	166 🗷
29	132	167
30	133	168
31	134	169
32	135	170
33	136	171
34	137	172
35	138	173
36	130	174
37	140	175 176 176 177 177 177 177 177 177 177 177
38	141	176 176
39	142 WOO	a
40	143	144

#### STATUS OF AMENDMENTS

On November 27, 1997, applicants submitted an amendment under 37 CFR 1.129(a) in response to the final rejection dated August 27, 1997. On May 14, 1997, applicants filed a supplementary response to the final rejection dated August 27, 1997. In response to these two submissions under 37 CFR 129(a), all pending claims except for claim 136 were finally rejected in the Final Action dated June 25, 1998. Applicants filed a notice of Appeal on December 28, 1998.

In the response to the Final Action dated June 25, 1998, applicants submitted the following Amendments after final rejection:

Amendments After Final Rejection:

November 25, 1998: Not entered (Advisory mailed 12/14/98 (Paper #68))

December 10, 1998: Not entered (Advisory mailed 2/25/99 (Paper 77C))

December 11, 1998: Not entered (Advisory mailed 2/25/99 (Paper 77A))

December 15, 1998 (2): [1.132 Declarations of Mitzi, Tsuei, Dinger and Shaw]

Entered (Advisory mailed 2/25/99 (Paper 77E))

December 15, 1998: [1.132 Declaration of James Leonard]

Not entered (Advisory mailed 2/25/99 (Paper 77B))

December 18, 1998: Entered (Advisory mailed 2/25/99 (Paper 77E))

December 22, 1998: Not entered (Advisory mailed 2/25/99 (Paper 77A))

December 27, 1998: Not entered (Advisory mailed 2/25/99 (Paper 77D))

June 14, 1999: Entered (Advisory mailed 7/8/99 (Paper #83))

June 15, 1999 (2): Entered (Advisory Actions mailed 7/8/99 (Paper #84 and #85))

<u>June 27, 1999</u>: Not entered (Advisory mailed 7/8/99 (Paper #86))

In response the five Advisory Actions dated February 25, 1999 were received by Applicants. In response to these Advisory Actions three Amendments of the Final Rejection were submitted on June 14, 1999; June 15, 1999 and June 15, 1999. By telephone conversation the Examiner told applicants that these would be entered.

On June 24, 1999, Applicants resubmitted the amendment after final dated December 27, 1998 which the Examiner told the undersigned attorney was not received.

#### SUMMARY OF THE INVENTION

A method of flowing a superconducting electrical current in a transition metal oxide having a  $T_c$  greater than 26°K and maintaining the transition metal oxide at a temperature less than said  $T_c$ .

In a more particular aspect of the method of the present invention the transition metal is copper.

In another more particular aspect of the method of the present invention the transition metal oxide has a perovskite-like or a perovskite-type crystal structure which is a perovskite or near perovskite structure.

In another more particular aspect of the present invention the transition metal oxide has a layered-like or layered-type structure.

In another more particular aspect of the present invention, the composition includes a transition metal, a rare or rare earth like element, an alkaline earth element and oxygen.

#### **ISSUES**

- 1. Are claims 24-26, 86-90, 96-135 and 137-177 supported by the priority document?
- 2. Are claims 24-26, 86-90, 96-135 and 137-177 anticipated under 35 USC 102(a) by the Asahi Shinbum article?
- 3. Are the claims 24-26, 86-90, 96-135 and 137-177 obvious under 35 USC 103(a) in view of the Asahi Shinbum article?
- 4. Are claims 24-26, 86-90, 96-113, 129-131, 134, 135, 139-142, 143-163, 164-166, 169, 170, and 174-177 not enabled under 35 USC 112, first paragraph.
- Are claims 86-87, 96-108, 115, 118, 120, 122, 123, 124, 129-135, 137-142,
   164-169, and 170-177 indefinite under 35 USC 112, second paragraph.

#### **GROUPING OF THE CLAIMS**

Each claim is appealed individually.

## **ARGUMENT**

# **Priority**

The Examiner has acknowledged applicant's claim for priority under 35 USC §119 in the parent application, Serial No. 08/053,307 filed April 23, 1993. The certified copy has been filed in parent application, Serial No. 08/053,307, filed on April 23, 1993 as paper no. 28. (References to the priority document herein are to the corresponding European Patent Application 275 343 A1 published on July 27, 1988.)

Applicants respectfully disagree with the Examiner that support is not found in that certified copy for the invention as presently claimed.

In this regard the Examiner states:

Applicants' arguments filed April 11, 1996, January 3, 1996 and September 29, 1996, paper numbers 53, 50 and 51, as well as the Affidavits filed September 29, 1995 and January 3, 1996, paper numbers 49 and 52, have been fully considered but they are not deemed to be persuasive. The applicants quote some passages out of the priority document and argue that the present claims are fully based on that document. Nevertheless, that priority document is not deemed to provide basis for the limitations found in the present claims.

In this passage the Examiner states that "Applicants' arguments ... are not **deemed** to be persuasive" and "[n]evertheless, that priority document is not **deemed** to provide basis for the limitations found in the present claims." Webster's Ninth New Collegiate Dictionary (Merriam-Webster Inc., Springfield, Mass. 1987) defines "deem" as a transitive verb meaning "to come to think or judge" and as an intransitive verb meaning "to have an opinion: believe." The examiner has used the intransitive form of the verb "deemed". The examiner has cited no statutory or case law authority which permits an examiner to object to a claim of priority based on the examiners "opinion" or "belief" that a priority document does not support applicant's claims. The Examiner must support a denial of a claim of priority based on what is actually stated in the priority document.

The examiner further states in support of the examiner's "opinion" or "belief" at page 3, paragraph 4.b;

i. The recitation of a "composition including a transition metal, a rare earth or rare earth-like element, an alkaline earth element, and oxygen", as found in claim 86 (lines 2-4). The certified priority document may provide basis for the formula RE<sub>2</sub>TM.O<sub>4</sub> at p. 2, para. 4, but the claimed composition is deemed to be much broader than that formula.

Applicants respectfully disagree. In the priority document, for example in the abstract, RE is a rare earth element, TM is a transition metal and O is oxygen. The priority document further states at Col. 2, lines 22-25 "the lanthanum which belongs to the IIB group of elements is in part substituted by one member of the neighboring IIA group of

elements...". Group IIA elements are the alkaline earth elements. The present specification teaches at page 11, lines 22-23, that RE stands for the rare earths (lanthanides) or rare earth-like elements. The "rare earth like element" act like a rare earth element in the superconductive composition. Thus a rare earth-like element is an equivalent of rare earth element. Similar language appears in the present specification at page 12 lines 6--8, "the lanthanum which belongs to the IIB group of elements is in part substituted by one member of the neighboring IIA group of elements...". Therefore, the priority document teaches a "composition including a transition metal, a rare earth or rare earth-like element, and alkaline earth. Applicants note that in the passage quoted above, the Examiner incorrectly states that applicants claim a composition. This is not correct. Applicants claim a method of flowing a superconducting current in a transition metal oxide. In the last sentence of the passage quoted above the Examiner incorrectly states "the claimed composition is deemed to be much broader than [the] formula" RE<sub>2</sub>TM.O<sub>4</sub>". The priority document is not limited to his formula. The composition taught by the priority document have variable amounts of oxygen, rare earth, rare earth-like and alkaline earth elements as is clearly shown in the abstract of the priority document.

# The Examiner further states:

ii. The limitation "non-stoichiometric amount of oxygen", as found in claim 6 and 86 (line 6). Basis may be seen for an oxygen deficit at p. 2, para. 4, but no such basis is seen for the more general limitation of "a nonstoichiometric amount of oxygen".

Applicants respectfully disagree. At Col. 3, lines 46-50 the priority document refers to applicants publication in Z. Phys. B - Condensed Matter 64 (1986) 189-193 which is incorporated by reference in the present specification at page 6, lines 7-10. (This article is referred to here in as Applicants' article.) This article states at page 190, left col., lines 13-14 "[t]his system exhibits a number of oxygen-deficient phases with mixed-valent copper constituents." The priority document has various general formulas such as at Col. 3, lines 40, "La₂xBaxCuO₄y x<<1 and y≥0." The abstract has a more generic formula. A stoichiometric compound has a fixed amount of each element that make up the compound. Since, the amount of oxygen is variable, the formula has nonstoichiometric amounts of oxygen. Therefore, the priority document teaches nonstoichiometric amounts or oxygen.

In attachment A of this appeal, there are copies of pages 245 and 225 of Inorganic Chemistry by Moeler, John Wiley & Sons, Inc. 1952 and a copy of page 70 of Fundamentals of Chemistry, A Modern Introduction by Brescia et al., Academic Press, 1966. Attachment A provides an explanation of the terms stoichiometric and nonstoichiometric. The documents in Attachment A support applicants position that the priority document teaches nonstochiometric amounts or oxygen.

#### The Examiner further states:

iii. The limitation "a composition exhibiting a superconductive state" is found in present claim 88, (line 2). Wherein the certified priority document

may provide basis for compositions of the formula RE<sub>2</sub>TM.O<sub>4</sub>, as discussed above, but "transition metal oxide" and "superconductive state" are deemed to be much broader than the formula RE<sub>2</sub>TM.O<sub>4</sub>.

Applicants respectfully disagree. The field of the invention of the priority document is "a new class of superconductors in particular components ..." and the title is "New Superconductive Compounds ...". Applicants' article which is referred to in the priority document states at page 190, left Col., lines 14-16 from the bottom "X-ray powder diffractograms ... revealed three individual crystallographic phases." In the conclusion at page 192 the article states "[t]he system consists of three phases, one of them having a metallic perovskite-type layer-like structure. The characterization of the new, apparently superconducting, phase is in progress." Thus the priority document supports the limitation "a composition exhibiting a superconductive state". The general formula  $RE_{2x}AE_xTM.O_{4y}$  x<0.3  $0.1 \le y \le 0.5$  and the more specific formula  $RE_2TM.O_4$  of the priority document is a composition; is a metal oxide; and is a transition metal oxide as recited in claims 24, 89 and 90. As noted above, the Examiner incorrectly implies that the priority document is limited to compounds having the formula  $RE_2TM.O_4$ .

#### The Examiner further states:

- iv. The limitation "a copper-oxide compound" is recited in claim 96 (line
- 6). The certified priority document may provide basis for compositions of the formula RE<sub>2</sub>TM.O<sub>4</sub> , as discussed above, but "a copper-oxide

compound" is not deemed to be equivalent to a composition of the formula RE<sub>2</sub>TM.O<sub>4</sub>. Basis is not seen in the certified priority document for "a copper oxide compound" with the breadth of the present claims.

Initially the Examiner incorrectly implies claim 96 is directed to a copper oxide compound. Claim 96 is directed to a "copper oxide composition consisting essentially of a copper oxide compound having a layer-type perovskite-like structure." Applicants respectfully disagree. The priority document recites numerous copper oxide compositions. It is noted that the Abstract of the priority document refers to "[t]he superconductive compounds are oxides of the general formula RE<sub>2-x</sub>AE<sub>x</sub>TM.O<sub>4-y</sub>, wherein RE is a rare earth, AE is a member of the group of alkaline earths or a combination of at least two members of that group, and TM is a transition metal, and wherein x < 0.3 and 0.1  $\le$ y  $\le$  0.5." This formula permits no alkaline earth and a varying amount of alkaline earth, rare earths and a varying amount of oxygen. At column 3, lines 20 and 35, there is recited "the Ba-La-Cu-O system" and at line 41 "La<sub>2-x</sub>Ba<sub>x</sub>CuO<sub>4-y</sub>x < 1 and y  $\leq$  0 and at line 44 teaches La<sub>1-x</sub>Va<sub>x</sub>CuO<sub>3-y</sub>. Thus the priority document provides support for a composition including a transition metal, a rare earth or rare earth-like elements, an alkaline earth element, an oxygen as found in applicants' claim, specifically claim 86. It is noted that at column 2, lines 13-19 the priority document states that "it is a characteristic of the present invention that in the compounds in question that the RE portion is partially substituted by one member of the alkaline earth group of metals, or by a combination of the members of this alkaline earth group and that the oxygen content is at a deficit." It is further noted that at

column 2, lines 20-23 it states that "for example, one such compound that meets the description given by this lanthanum copper oxide La₂CuO₄ in which the lanthanum which belongs to the IIIB group of the elements is in part substituted by one member of the neighboring IIIA group of elements."

The priority document at column 3, line 6 recites Ti as a transition metal. It is noted that in claim 1 of the priority document, claim 1 recites the structure RE<sub>2-x</sub>AE<sub>x</sub>TM.O<sub>4-y</sub> wherein TM is a transition metal. Claim 2 therein recites copper as the transition metal. Claim 3 therein recites nickel as the transition metal. Claim 8 therein recites chromium as the transition metal. Consequently, a broader class of transition metals other than copper is supported by the priority document.

It is clear from the quoted sections of the priority document that the priority document clearly supports a much broader composition than the Examiner is claiming that it does, and that the priority document, in fact, does support applicant's claims.

As noted above, the general formula of the priority document is much broader than the formula RE<sub>2</sub>TM.O<sub>4</sub> which the Examiner incorrectly states the priority document is limited to. The quantity of oxygen, the rare earth element and of an alkaline element is variable and the transition metal is not limited to copper. Consequently, the term "a copper-oxide compound" is adequately supported by the priority document.

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The Examiner further states:

v. The limitation to the effect that "the copper oxide compound includes (including) at least one rare-earth or rare-earth-like element and at least one alkaline-earth element", as recited in claim 103 (lines 6 and 8). The certified priority document may provide basis for compositions of the formula RE<sub>2</sub>TM.0<sub>4</sub>, as discussed above, but basis is not seen for the more general limitation of "a copper-oxide compound" with a rare-earth (like) element and an alkaline earth element.

Applicants respectfully disagree. The second line of the abstract gives the general formula "RE2\*AE\*TM.O4\*y x<<0.3 and  $0.1 \le y \le 0.5$ ." In claim 1 of the priority document  $y \le 0.5$ . Claim 2 recites RE is lanthanum and TM is a copper. Claim 3 recites RE is cerium and TM is nickel. Claim 4 recites RE is lanthanum and TM is nickel. Claim 8 recites RE is lanthanum and TM is chromium. Claim 9 recites RE is neodymium and TM is copper. Applicants' claim 103 recites "the copper-oxide compound including at least one rare-earth or rare-earth-like element and at least one alkaline-earth element". The priority document clearly supports this recitation. Applicants, as stated above, respectfully submit the Examiner is misrepresenting the priority document which refers throughout and, in particular, in the Abstract to "the general formula RE2\*AE\*EM.O4\*y as stated above which includes a copper-oxide as stated above. The Examiner further states in the passage quoted above "but basis is not seen for the more general limitation of 'a copper-oxide compound' with a rare-earth (like) element and in alkaline earth element." It is noted that in the priority document, claim 2 refers to lanthanum as

the rare earth; claim 3 refers to cerium as the rare earth; claim 5 refers to barium as a partial substitute for the rare earth; claim 6 refers to calcium as a partial substitute for the rare earth; claim 7 refers to strontium as a partial substitute for the rare earth and claim 9 refers to neodymium as the rare earth. Clearly, the priority document uses barium, calcium and strontium. Consequently, the priority document supports the term rare earth-like since in includes elements (e.g. barium, calcium and strontium) other than those commonly referred to as the rare earth elements [which are elements 57-71] which satisfy the teaching of the priority document and of the present application. The Abstract of the priority document refers to "AE as a member of the alkaline earth or a combination of at least two members of that group". Consequently, the priority document clearly supports an alkaline earth element.

#### The Examiner further states:

vi. The limitation to the effect that "the copper-oxide compound includes at least one element (oxygen) in a nonstoichiometric atomic proportion", as found in claim 101 (lines 2 and 3), 102 (lines 2 and 3), 107 (lines 2 and 3), and 108 (lines 2 and 3). Basis may be seen for an oxygen deficit as discussed above, but no such basis is seen for the more general limitation of "a nonstoichiometric atomic proportion".

Applicants disagree for the same reasons given above for why the priority document supports "nonstoichiometric amount of oxygen".

The Examiner further states:

vi. The limitation as to "the effectively-zero-bulk-resistivity intercept temperature  $T_{\rho=0}$ , as found in claim 103 (lines 13, 6 and 17). The critical temperature,  $T_c$ , is discussed throughout that certified priority document, but not  $T_{\rho=0}$ .

Applicants respectfully disagree.  $T_{\rho=0}$  is the temperature at which the bulk resistively is about zero. T<sub>c</sub> is the critical temperature or the temperature above which superconductivity does not exist. The priority document refers to applicants' article of which Figures 1,2 and 3 are the same figures as Figures 2, 3 and 4 of the present application. At page 22, lines 19-24, the present specification refers to Figure 4 of the specification stating "[i]ts resistivety decreases by at least three orders of magnitude, giving evidence for the bulk being superconcucting below 13 K with an onset around 35 K, as shown in FIG. 4 on an expanded scale." When a superconductor is totally superconductive the resistivety,  $\rho$ , is zero. The temperature at which this occurs is  $T_{\rho=0}$ . Applicants' article, (and thus the priority document), at page 191, right column, in referring to Fig. 1 thereof states "[u]pon cooling from room temperature, the latter exhibit a nearly linear metallic decrease of  $\rho(T)$ , then a logarithmic type of increase, before undergoing the transition to superconductivity." And in the sentence bridging pages 191-192 "[t]herefore, under the above premises, the peak in  $\rho(T)$  at 35 K, observed ... has to be identified as the start to superconductive cooperative

phenomena." And Applicants' article at page 192, left column, states "[u]pon cooling below T<sub>c</sub> ... the bulk resistively gradually drops to zero by three orders of magnitude, for sample 2( Fig. 1)" From these statements in applicants article (which is referred to in the priority document) it is clear that the language objected to by the examiner is supported in the priority document.

In response to Applicants' arguments filed March 7, 1997 (#59) the Examiner states "they have been fully considered but not found to be persuasive".

The Examiner states:

i. The applicants quote portions out of the priority document and assert that those quoted sections "clearly (support) a much broader composition than the Examiner is claiming it does, and that the priority document, in fact, does support applicant's (sic) claim 86." The fact remains, nevertheless, that the priority document refers to the general formula RE<sub>2</sub>TM.0<sub>4</sub> in which the rare earth element (RE) may be partially substituted with a Group IIA metal. That disclosure in the priority document does not provide support for the broader limitations of the present claims, which do not limit the invention to that general formula.

As state above the Examiner incorrectly states that the priority document is limited to formula RE<sub>2</sub>TM.0<sub>4</sub>. This is clearly incorrect.

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The Examiner further states:

ii. The applicants argue that the disclosure of varying amounts of oxygen in the priority document provides support for earlier priority for the term "non-stoichiometric amount of oxygen". Again, however, that disclosure in the priority document does not provide support for the broader limitations of the present claims, which do not limit the invention to those varying amounts.

It is not clear what the Examiner is trying to say in the last sentence of this quoted passage. The general formula in the Abstract of the priority document, RE<sub>2-x</sub>AE<sub>x</sub>TM.O<sub>4-y</sub>, has the atomic amount of O varying from 0 to 4 independent of the atomic amount of RE, AE and TM. Thus the amount of O must have non-s toichiometric values. The Examiner's comments in the passage above are clearly in error.

The Examiner further states:

iii. The applicants urge that the disclosure in the priority document of the formula RE<sub>2</sub>TM.0<sub>4</sub> provides support for their limitations of "transition metal", "copper-oxide compounds", "rare earth or rare earth-like elements", and "alkaline earth element". Again, however, that disclosure in the priority document does not provide support for the broader

limitations of the present claims, which do not limit the invention to that formula.

The Examiner again incorrectly states that the priority document is limited to the formula RE<sub>2</sub>TM.O<sub>4</sub>. As stated above the examiner is clearly in error. The priority document supports a much broader formula then RE<sub>2</sub>TM.O<sub>4</sub>. As shown above, the priority document clearly supports applicants' claim limitations "transition metal", "copper-oxide compounds", "rare earth or rare earth-like elements" and "alkaline earth elements".

#### The Examiner further states:

iv. The applicants further "assume that the Examiner agrees with applicant's (sic) statements in their prior response in that the concept of the intercept temperature is well known in the prior art and can be included in claim 103." No basis is seen for that assumption. As noted in the previous Office Action and repeated above, the term "T $_{\rho=0}$ " is not found in the priority document. Well known or not, there is no basis for that term in the priority document.

For the reason given above the priority document clearly supports the term " $T_{\rho=0}$ ". Although this particular symbol is not used in the priority document, the priority document clearly shows that as temperature is decreased the resistively of a

superconductor begins to drop in the value at the critical temperature  $T_c$  and goes to zero at another temperature, that is  $T_{\rho=0}$ . This symbol is just a short hand notation for that temperature. This property of superconducting materials is well known prior to applicants filing date, in fact that is what is meant by the term superconductor which is a material for which  $\rho=0$  for temperatures less than a certain temperature, i.e.,  $T_{\rho=0}$ . It is also well-known that: "[i]n the ideal case the resistance vanishes completely and discontinuously at a transition temperature. Ts ... Actually, the resistance temperature curve does fall more sharply the more specimen is like a single crystal ... [T]he drop always occurs in a measurable temperature range ..." (Theory of Superconductivity, M. von Laue, Academic Press, Inc., 1952) (See Appendix C of Applicants' response dated September 25, 1995). Moreover, the priority document at column 1, the first sentence of the Background of the Invention states "[s]uperconductivity is usually defined as the complete loss of electrical resistance of a material at a well defined temperature". That temperature is symbolically represented as  $T_{\rho=0}$ .

# **CLAIM REJECTIONS - 35 USC § 102**

Claims 24-26, 86-90, 96-135, and 137-177 have been rejected under 35 U.S.C. § 102(a) as being anticipated by Asahi Shinbum, International Satellite Edition (London), November 28, 1986 (hereinafter, "the Asahi Shinbum article").

The Examiner is stating that everything within applicants non-allowed claims is found in the Asahi Shinbum article. All of Applicants' non-allowed claims are dominant to (or generic to) the one allowed claim, claim 136. Thus by stating that all the non-allowed claims are anticipated, the Examiner is stating that the portion of each non-allowed claim which does not overlap the allowed claim is taught in the Asahi Shinbum article. This means that a person of skill in the art needs nothing more that what is taught in the Asahi Shinbum article to practice that part of each of Applicants non-allowed claims which does not overlap Applicants' allowed claim.

The only parts of the Asahi Shinbum article which are relevant to applicants claim are in the first paragraph:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition has been found. The possibility of high  $T_c$  - superconductivity has been reported by scientists in Switzerland this spring. The group of Prof. Shoji TANAKA, Dept. Appl. Phys. Faculty of Engineering at the University of Tokyo confirmed in November, that this is true.

in the second paragraph:

The ceramic newly discovered, is an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties. Prof. Tanaka's laboratory confirmed that this material shows diamagnitism (Meisner effect) which is the most important indication of the existence of superconductivity.

The Swiss scientist are the inventors of the present application. Thus this clearly refers to applicants work which was reported in Applicants article. These passages say that Prof. Tanaka confirmed applicants work. The newly discovered ceramic referred to in the article is the ceramic reported on in Applicants' article. The present applicant was filed less than one year after the publication of applicants' article. This article is a disclosure of Applicants' own invention and cannot be used as a reference against the present application.

Since the present application was filed within one year of Applicants' article, Applicants' article is not a reference as to them. Thus the only portion of the Asahi Shinbum article relevant to Applicants' claims is:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition ... is an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties.

Since the Asahi Shinbum article refers to " $T_c$  of 30K" and since each of Applicants claims recites  $T_c > 26$  °K, the Asahi Shinbum article alone cannot anticipate  $T_c > 26$  °K since to be an anticipation a reference must contain all the limitations of the claim it is said to anticipate. Also, the Asahi Shinbum article provides no teaching of how to made the "new ceramic". A reference which does not provide a method of making a composition cannot anticipate a claim to the composition and thus to a use of that new composition. Also, the Asahi Shinbum article has no specific embodiment of the new composition. Thus it cannot anticipate under 35 USC 102.

# Applicants' Article Was in the US in Ready To File Form

The issue involved here is straight forward. A third party reproduced and reported that fact in a written publication before of Applicants' filing date. The work of Applicants was reported in a written description published before the publication of the third party. Applicants filed the present application within one year of their publication, but after the third party publication. If Applicants did not file the present applicant within one year of the date of Applicants' article, Applicants' article would be a valid reference under 35 USC 102(a). But since Applicants filed the present application within 1 year of Applicants' article, it is not a reference under 35 USC 102(b).

35 USC 100 states the term "invention" means invention or discovery.

35 USC 102(a) states "[a] person shall be entitled to a patent unless ... the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicants for patent."

Applicants invention or discovery was on or before April 17, 1986 which is the date Applicants submitted Applicants' article to Z.Phys.B. That article was published in September, 1986. Therefore, Applicants invented their invention prior to the date of the Asahi Shinbum article, November 28, 1986. Evidence submitted proving that applicants conception was in the United States at applicants direction prior to Nov. 28, 1986 is discussed below. In addition, the following evidence shows that Applicants' article was in this country in possession of IBM, the assignee, prior to the date of the Asahi Shinbum article. Attachment K of Applicants' response dated December 27, 1998, page 1, is a copy of the front cover of Zeitschrift Fur Physik B Condensed Matter Vol. 64 which contains Applicants' article ( pp 189-193) which is referred to and incorporated by reference at page 6, lines 6-10, of Applicant's specification. Applicants state at page 6 of the specification that Applicants' article is "[t]he basis or our

invention". This page bears in the upper right the date stamp of the IBM Research Library bearing the date of Sept. 18, 1986. Page 2 of Attachment K of Applicants' response dated December 27, 1998, is an enlarged view of the upper right corner showing the date stamp. Thus the assignee of the present invention, IBM, who was the employer of the inventors at the time of the conception of the invention, had in its possession in the United States, prior to the date of the Asahi Shinbum, a copy of the article which Applicants state forms the basis of their invention. Thus IBM had in its possession in the United States a written description of applicants' invention in "ready to patent form" (as defined by the United States Supreme Court in Pfaff v. Wells 48 USPQ 2d 1641 decided November 10, 1998) prior to the date of the Asahi Shinbum article. The US Supreme Court held that "reduction to practice" is not needed to establish a date for invention. The court stated "[t]he statute's only specific reference to that term is found in §102(g), which sets fort the standard for resolving priority between two competing claimants to a patent." Since §102(g) is not applicable here, "diligence" and "reduction to practice" are not required. Applicants article in Zeitschrift Fur Physik "is proof that prior to [the date of the Asahi Shinbum article applicants have] prepared drawings or other descriptions of the invention that were sufficiently specific to enable a person skilled in the art to practice the invention." The Asahi Shinbum article sates that applicants' work was reproduced, by others, thus applicants article was sufficiently specific for a person of skill in the art to practice applicants' invention. Also, as stated in Applicants' response dated December 18, 1998, more than 5,200 articles refer to applicants article showing that applicants enabled the field of high T<sub>c</sub> superconductivity. Thus the Asahi Shinbum article is not a valid §102(a) reference against Applicants' claimed invention.

### The Examiner states:

ii. The applicants assert that the Asahi Shinbum article reports a third party's confirmation of their original discovery. That assertion appears to be correct, but the article still is deemed to be prior art under 35 USC 102(a).

(1) It should be noted again, however, that the applicants' discovery was not originally made in this country- and that they cannot show an earlier date than December 1986 for their invention in this country. The Asahi Shinbum article was published on November 28, 1986.

Applicants disagree with the Examiner. Applicants note that the Examiner acknowledges that the Asahi Shinbum article "confirms [Applicants'] original discovery." 35 USC 102(a) does not require applicants to show a date of invention in this country prior to the Asahi Shinbum article to avoid the Asahi Shinbum article being prior art. 35 USC 102(a) states "A person shall be entitled to a patent unless ... the invention was ... described in a printed publication in this or a foreign country, before the invention thereof by the application for patent." 35 USC 100 does not include reduction to practice in this country or conception in this country as part of the definition of invention. Only 35 USC 102(g) includes the language "reduction to practice" and "invention in this country". If Congress intended a reduction to practice to be necessary for a patent applicant to show that they invented their invention before the date of the printed publication, Congress would have included such language in the 35 USC 102(a). Also, if Congress intended that invention in this country was to be necessary for a patent applicant to show that they invented their invention before the date of the printed publication, Congress would have included such language in 35 USC 102(a).

In the final rejection the Examiner states:

a. As discussed in paper no. 20 of the ancestral application,
07/053,307, it is not fully clear to what exact date applicants are entitled.
Based on the record, nonetheless, that date would appear to be no later

than around December 13, 1986, the date samples were tested in the US to show superconductivity. See MPEP 715 et seq. The Asahi Shinbum article was published on November 28, 1986.

b. The reference confirms superconductivity in an oxide compound of La and Cu with Ba having a structure of the so-called perovskite structure. Although the reference fails to teach use of the testing of zero resistance for confirming superconductivity, it inherently must have been used because it is one of two methods used for testing for superconductivity (the other being diamagnetism). Accordingly, the burden of proof is upon the applicants to show that the instantly claimed subject matter is different from and unobvious over that taught by this reference."

In response to applicants' arguments filed April 11, 1996, January 3, 1996, and September 29, 1995, paper nos. 53, 50, and 51, as well as the Affidavits filed September 29, 1995 and January 3, 1996, paper nos. 49 and 52, the Examiner states they "are not **deemed** to be persuasive", that is it is the Examiner's **opinion** that they are not persuasive.

The Examiner cites In re Brown, 173 USPQ 685, 688; In re Best, 195 USPQ 430; and In re Marosi, 218 USPQ 289, 293 to support his rejection. These decisions are not directed to whether a reference is a valid §102(a) reference and are thus not relevant to this issue.

The Examiner is using Asahi Shinbum as a reference under 35 USC §102(a). Applicants disagree that this is proper since to do so does not permit applicants the one

year period provided under 35 USC §102(b) to file a US application after their own publication. The one year period permitted applicants to file the present application up to September 1987. The date of the Asahi Shinbum article November 28, 1986 is after the date of applicants' publication, but before the end of the one year.

Applicants believe that the Examiner has incorrectly applied 35 USC §102(a). The Court of Custom and Patent Appeal in In re Katz 215 USPQ 14, 17 states that:

It may not be readily apparent from the statutory language that a printed publication cannot stand as a reference under §102(a) unless it is describing the work of another. A literal reading might appear to make a prior patent or printed publication 'prior art' even though the disclosure is that of the applicant's own work. However, such an interpretation of this section of the statute would negate the one year period afforded under §102(b) during which an inventor is allowed to perfect, develop and apply for a patent on his invention and publish descriptions of it if he wishes.

Thus, one's own work is not prior art under §102(a) even though it has been disclosed to the public in a manner or form which otherwise would fall under §102(a). Disclosure to the public of one's own work constitutes a bar to the grant of a patent claiming the subject matter obvious therefrom only when the disclosure occurred more than one year prior to the date of the application, that is, when the disclosure creates a one-year time bar, frequently termed a "statutory bar," to the application under

§102(b). As stated by this court in In re Facius, 56 CCPA 1348, 1358, 408 F.2d 1396, 1406, 161 USPQ 294, 302 (1969), "But certainly one's own invention, whatever the form of disclosure to the public, may not be prior art against oneself, absent a statutory bar." [Emphasis in original].

The Asahi Shinbum article states in the first paragraph, "The possibility of high T, superconductivity has been reported by scientists in Switzerland this spring". The "scientists in Switzerland' are the inventors of the present application. Applicants' invention was reported in Applicants' article which was submitted for publication in the Spring of 1986. The Asahi Shinbum article only reports the work of applicants and that it was reproduced by Prof. Tanaka. This article is a disclosure of applicants' "own invention" and cannot be used as a reference. Therefore, the Examiner is in error in rejecting Applicants claims 24-26, 86-90, 96-135 and 137-177 under 35 USC §102(a) as anticipated by Asahi Shinbum and under 35 USC §103 as obvious over Asahi Shinbum.

In regard to the two-year grace period under a prior statute corresponding to 35 USC §102(b) the U.S. Supreme Court in Andrews v. Hovey, 123 US 267 (1887) states that:

"The evident purpose of the section was to fix a period of limitation which should be certain, and require only a calculation of time, and should not depend upon the uncertain question of whether the Applicant had consented to or allowed the sale or use. Its object was to require the

inventor to see to it that he filed his application within two years from the completion of his invention, so as to cut off all question of the defeat of his patent by a use or sale of it by others more than two years prior to his application, and thus leave open only the question of priority of invention. The evident intention of congress was to take away the right which existed under the act of 1836 to obtain a patent after an invention had for a long period of time been in public use, without the consent or allowance of the inventor; it limited that period to two years, whether the inventor had or had not consented to or allowed the public use." (Emphasis added)

From this quote from Andrews v. Hovey, it is evident that the use or sale by others prior to filing a patent application by the inventor does not cut off the inventors right to obtain a patent so long as the inventor files the application within the statutory period which was 2 years at the time of the Andrews v. Hovey decision and is now 1 year under 35 USC 102(b). (Applicants note that the U.S. Supreme Court cited Andrew Hovey with approval in Pfaff v. Wells.) Thus Prof. Tanaka's reproducing of Applicants' results reported in Applicants' article and the reporting of this in Asahi Shinbum article does "not cut off [Applicants'] right to obtain a patent" since Applicants have filed the present application within one year of the date of publication of Applicants' article. Applicants note that the Supreme Court says that "the consent or allowance of the inventor" is not a factor in determining whether "a use" by another cuts off the one year period under §102(b).

The Patent Office Board of Appeals in Ex parte Powell and Davies, 37 USPQ

285 states in regard to the publication of applicant's foreign patent application before

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the filing of a U.S. application on October 5, 1936 on an invention described in the foreign patent application that:

The Examiner has also rejected the claims on the printed specification of Applicants' own British application which appears from this record to have been published on August 27, 1936. We know of no authority for such a rejection. Neither section 3886 nor section 4887 R.S. warrants the rejection. Obviously, the publication could not have a date prior to Applicants' invention. There is no statute that requires an Applicant to make his invention in this country.

Therefore, Applicants of the present invention can rely on their publication in Zeitschrift for Physik as evidence of their invention. Applicants note that the Board states that the statute does not require Applicants to make the invention in this country to get the benefit of the one year period under 35 USC §102(b). Therefore, the date of Applicants' invention is as least as early as the date of Applicants' publication which is before the date of the Asahi Shinbum article.

The Patent Office Board of Appeals in Ex parte Powell and Davies, 37 USPQ 285, 286 further states:

The Commissioner indicates in Ex parte Grosselin that the Examiner should consider whether the German patent was derived from Applicant and was in effect nothing more than a printed publication of Grosselin's invention.

The Asahi Shinbum article states in the first paragraph:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition has been found. The possibility of high  $T_c$  - superconductivity has been reported by scientists in Switzerland this spring. The group of Prof. Shoji

TANAKA, Dept. Appl. Phys. Faculty of Engineering at the University of Tokyo confirmed in November, that this is true.

The "scientists in Switzerland" are the inventors of the above-identified application. The Asahi Shinbum article only reports the work of Applicants and that it was reproduced by Prof. Tanaka. This article is a disclosure of Applicants' "own invention" and clearly in the words of the Board in Ex parte Powell and Davies, "was derived from [Applicants] and [is] in effect nothing more than a printed publication of [Applicants'] own invention and cannot be used as a reference".

The Patent Office Board of Appeals in Ex parte Lemieux 148, 140 states that: Finally, we believe that our holding is consistent with decisions in interference practice wherein, even though in the usual case a party may not establish a priority date of invention by reference to activity in a foreign country, yet in an originality case where a party is seeking to prove that the other party derived from him so that there is only a single original inventor, he may be permitted to prove derivation by reference to activity abroad. ... By analogy, in the present case appellant has demonstrated that he is the single original inventor, there being no adverse party.

Following this decision it is clear from the Asahi Shinbum article that Applicants are the "single original inventor" and that the Asahi Shinbum article is "derived" from Applicants and that Professor Tanaka's work reported in the Asahi Shinbum article is "derived" from Applicants.

In In re Mathews 161 USPQ 276 (CCPA 1969) a patent to Dewey was cited under 35 USC 102(e) against the application of Mathews. The Dewey patent disclosed but did

not claim the invention claimed in the Mathews application. The claimed invention in the Mathews application was a circuit which Dewey disclosed to comply with 35 USC 112 requirements. Mathews submitted Dewey's affidavit under 37 CFR 132 which stated that Mathews disclosed to Dewey the circuit which Dewey described but did not claim. The CCPA held that Dewey was not a reference under 35 USC 102(e) against Mathews application stating 162 USPQ 276, 278:

It necessarily follows that Dewey may not be relied upon to defeat

Mathews' application since Dewey's disclosure, in view of the facts

established in the record, is not inconsistent with the novelty of Mathews'

claimed invention, That is, on the record here, Dewey derived his

knowledge from Mathews who is "the original, first, and sole inventor."

Following In re Mathews, it is necessary follows that the Asahi Shinbum article cannot be relied upon under 35 USC 102(a) to defect the present application since the Asahi Shinbum article states that Prof. Tanaka derived his knowledge from applicants who are the original, first, and sole inventors.

In re Mathews is directed to a reference under 35 USC 102(e) and not under §102(a). But this does not matter since under §102(e) an issued patent (which corresponds to a printed publication under §102(a)) is a reference as of the filing date (which corresponds to the publication date of a printed publication under §102(a)) and not the publication date (the issue date) of the §102(e) cited patent. Also, if the patent

cited as a §102(e) reference had issued prior to the filing date of the applicant in In re Mathews, it would have been a §102(a) reference. Thus the rational of In re Mathews should apply to a reference cited under §102(a). The Asahi Shinbum article states that Prof. Tanaka derived his knowledge from Applicants article and that Prof. Tanaka reproduced Applicants' work reported in Applicants' article and thus Applicants are the original, first and sole inventor.

In response to applicants comments on the cited decisions, the Examiner states, "The applicants cite four decisions which do not directly apply to the present facts."

Applicants disagree.

In regards to In re Katz the Examiner states:

(a) The In re Katz decision held that an applicant may overcome an article as 35 USC 102(a) prior art by showing that the applicant was a co-author and that the other co-authors were under the direction and control of the applicant. Here, however, the applicants were neither co-authors in the Asahi Shinbum article nor did they exercise direction and control over the work reported in that article.

Applicants disagree. The Examiner does not cite the text of In re Katz to support this interpretation of In re Katz. In fact, In re Katz does not support the Examiner's position. In In re Katz an article co-authored by the patent applicant was cited against

the applicants' patent application under 35 USC 102(a). The application was filed less than one year after the article. In determining whether the article was prior art under 35 USC 102(a), the CCPA states "[i]t may not be readily apparent from the statuary language that a printed publication can not stand as a reference under §102(a) unless it is describing the work of another." 215 USPQ 14, 17. The inventor submitted a declaration stating that he was the sole inventor of the subject matter described in the article and that the other authors were students working under his direction. The CCPA concluded that "The applicant's declaration is sufficient in this case to overcome the rejection" under 35 USC 102(a). 215 USPQ 14, 18. There is no evidence of record that the Asahi Shinbum article describes any invention other than those of Applicants. In this regard the CCPA further states:

As an initial matter, we hold that authorship of an article by itself does not raise a presumption of inventorship with respect to the subject matter disclosed in the article. Thus, co-authors may not be presumed to be coinventors merely from the fact of co-authorship. On the other hand, when the PTO is aware of a printed publication, which describes the subject matter of the claimed invention and is published before an application is filed (the only date of invention on which it must act in the absence of other proof), the article may or may not raise a substantial question whether the applicant is the inventor. For example, if the author (whether he is the applicant or not) specifically states that he is describing the work of the applicant, no question at all is raised. The content and nature of the printed publication, as well as the circumstances surrounding its publication, not merely its authorship, must

be considered. (Emphasis added).

It is clear form this passage that where the authors of an article are not the inventors of an invention described therein, the article is not necessarily a 102(a) reference. The above passage states "if the author (whether he is the applicant or not) specifically states that he is describing the work of applicants, no question at all is raised", that the article is not a reference under § 102(a). The Asahi Shinbum article clearly states that Prof. Tanaka reproduced applicants work reported in Applicants' article. There can be no question that the Asahi Shinbum article is not a reference under § 102(a). Moreover, "the content and nature" of the Asahi Shinbum article "as well as the circumstances surrounding the publication" clearly show that it is describing Prof. Tanaka's reproduction of Applicants' work. A published article is an invitation to all readers to reproduce an verify the work reported. Thus the Asahi Shinbum article describes no invention other than that of Applicants.

In regards to Andrews v. Hovey the Examiner states:

(b) The Andrews v. Hovey" decision involved a grace period which is now codified in 35 USC 102(b). The present case involves a printed publication as prior art under 35 USC 102(a).

The Examiner's comments miss the point of Andrews v. Hovey. As stated above, this case clearly says that "the use" by others prior to filing of the patent application by the inventors (which would be a §102(a) reference) does not cut off the right of the inventors to obtain a patent on the application filed within the statutory period under

§102(b) form the inventors own printed publication. The Asahi Shinbum article describes "the use" of Applicants' invention by Prof. Tanaka. If such a use in the United States would not be prior art under §102(a), it is not possible for a printed publication describing such a use in a foreign country to be prior art under §102(a).

In regard to Ex parte Powell and Davies:

(c) The Ex parte Powell and Davies" decision held that an applicant's own foreign patent which issued within the grace period cannot be used against him or her.

The Examiner's comments miss the point of Ex parte Powell and Davies which explicitly states that "[t]here is no statute that requires an Applicant to make his invention in this country". Applicants initially made there invention in Zurich, before the date of the Asahi Shinbum article. Their results were published prior to the Asahi Shinbum article in Applicants' article which was in possession of the assignee of the present invention in the US prior to the date of the Asahi Shinbum article. Also, in Ex parte Powel and Davies the Applicant's British application was published before their application was filed in the United States and it was still not a valid §102(a) reference.

In regard to Ex parte Lemieux, the Examiner states

The Ex parte Lemieux" decision applied that reasoning to an applicant's own article published in another country. Again, the present applicants had no part in the writing of the Asahi Shinbum article.

Again the Examiner's comments miss the point of Ex parte Lemieux which states "in an originality case where a party is seeking to prove that the other party derived from him so that there is only a single original inventor, he may be permitted to prove derivation by reference to activity abroad". Here the Asahi Shinbum article says that Prof. Tanaka derived his work from the work of Applicants and thus the Asahi Shinbum article is not a reference against the present application.

In regard to In re Mathews the Examiner states:

"In re Mathews, 161 USPQ 276, 277-279 (CCPA 1969), held that an applicant may overcome a patent as prior art under 35 USC 102(e) with evidence that the applicant provided the knowledge for the disclosure in that patent. By contrast, the present facts involve prior art under 35 USC 102(a) with a publication date before the invention was in this country."

In the present Application the Asahi Shinbum article acknowledges that

Applicants provided the knowledge to Prof. Tanaka to reproduce Applicants work which
is described in Applicants' article. And since the Asahi Shinbum article refers to

Applicants' discovery, as stated above, for this purpose a §102(a) reference is
equivalent to a 102(e) reference.

The Examiner further states:

(3) The present facts may raise a novel issue of law." The applicants were the first to develop the presently claimed invention, but the earliest

date they can show that invention in this country is December of 1986." The Asahi Shinbum article was published in November of 1986 and describes the development of superconductivity with an oxide of La, Ba, and Cu having a perovskite structure by a third party, but that article apparently indicates that the third party was confirming the discovery of the present applicants. Notwithstanding the possible uniqueness of the present facts, however, the Asahi Shinbum article still is deemed to be prior art under 35 USC 102(a), which the applicants have not been able to overcome with a showing of an earlier date in this country or a showing of their direction and control over the work done by that third party.

35 USC §102(a) and the cited case do not require work at an earlier date in this country to overcome reference cited under §102(a). Also, §102(a) and the cited case law do not require a showing that the Applicants exercised direction and control over Prof. Tanaka or the author of the Asahi Shinbum article. In fact, 35 USC §102 and the cited cases require a contrary result as Applicants have shown above. Notwithstanding, Applicants have shown that their conception was in this country in ready to file form, in the possession of the assignee of the present invention, prior to the date of the Asahi Shinbum article.

Applicants have argued that if one would follow the rationale of the Examiner, that is, if an applicant publishes an article and some other third party reports that same result prior to applicant's filing of a patent application which is subsequently filed within one year of applicant's own publication, the reporting of applicant's work by the third party would be prior art against applicant's application. Such a result would

deny (the applicant) the one year grace period provided under 35 USC 102(b). The Examiner dismisses this argument saying

"applicants" argument is duly noted, but again, it is further noted that the reference is prior art under 3 5 USC 102(a). The reference is not just a republication of the applicants' article. Instead, the reference is the reporting of someone else's work which confirms the applicants' work. The applicants also are not able to show a priority date which pre-dates the publication of that reference".

The Examiner is ignoring the fact that the Asahi Shinbum article and the work of Prof. Tanaka reported on therein was derived from Applicants. The cases cited above clearly state that when a third party derives their knowledge from an applicant, the third party's knowledge, for use or for publication of the information is not prior art against such an applicants' patent application.

## **Applicants Have Proven They Can Swear Behind the Reference**

Even though, as stated by the U.S. Supreme Court in Pfaff v. Wells, it is not necessary to show anything more than a conception to establish a date of invention under all sections of 35 USC 102 other than §102(g). Applicants have proven by facts that the conception of their invention was in the United States at their direction prior to the date of the Asahi Shinbum article, November 28, 1986, and applicants have proven that they were diligent from prior to the date of the Asahi Shinbum article by instructing coworkers in the United States until December 3, 1986 which is the date the Examiner

believes is the earliest date of Applicants reduction to practice in the United States. (For the reasons of record applicants believe that they have shown that their invention was reduced to practice in the United States prior to the date of the Asahi Shinbum article). The examiner has not rebutted applicants proof that applicants conception was in the United States at their direction prior to the date of the Asahi Shinbum article and the Examiner has not denied that applicants have proven that they were diligent by instructing coworkers in the United States from a time prior to the date of the Asahi Shinbum article until the date the Examiner believes is the date of Applicants' date of reduction to practice in the United States. The details of Applicants' proof are discussed at pages 22, line 8 to page 24, last line, of applicants' Substitute Amendment dated March 6, 1997. That argument is reproduced in detail below.

The Examiner's response to Applicants' proof is at page 19, paragraph d. ii of the final rejection "[t]he applicants further urge that they have shown clear diligence from before November 28, 1996 until actual reduction to practice at or around December 3, 1986. Nevertheless, the actual reduction in this country is deemed to have occurred on December 3, 1986, which is after the publication date for the reference." (As stated above the Examiner is in error that Applicants have to prove reduction to practice in this country before the date of the Asahi Shinbum article to avoid it as a §102(a) reference.)

Willson v. Sherts 81 F 2d 775, 28 USPO 379 (CCPA 1936) held (in an interference) that an inventor who conceives an invention outside of the United States

gets the benefit of the date that a third party, to whom the invention is disclosed, brings the conception into the United States (28 USPQ 379, 381) and that acts in this country done on behalf of the inventors can be used to show diligence to reduction to practice in the United States (28 USPQ 379, 383). Thus, the rejections of Applicants' claims under 35 USC 102 and 103 over the Asahi Shinbum article should be withdrawn.

It is noted that in the declaration of co-inventors J. G. Bednorz and K. A. Mueller dated March 21, 1988, mailed into the patent office on June 22, 1988 at paragraph 3, states "On approximately October 16, 1986, we gave Praveen Chaudhari ... six samples of the high temperature superconductive ceramic oxide materials that we had described in our aforementioned Z Physik B. publication. Praveen Chaudhari brought these samples back to the U.S. when he returned after visiting with us on or about October 16, 1986." This is evidence that these samples are brought into the United States on or about October 16, 1986. When these samples came into the United States, since they were inherently superconductive as claimed, the invention was essentially reduced to practice in the United States on that date. It is further noted that the Declaration of Alexis P. Malozenoff signed March 30, 1988 states at paragraph 3, "On or about November 15, 1986, Richard Greene and I traveled to Baltimore for a magnetics conference. During our travel to Baltimore, we discussed Greene's ongoing experiments in high T<sub>c</sub> superconducting samples which he said had been received from Bednorz and Mueller." This is clear evidence that by November 15, 1986, superconducting samples fabricated by applicant's were being measured in the United States. These samples were inherently superconducting and, consequently,

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established the reduction to practice in the United States as of that date. The Declaration of Cheng-Chung John Chi dated March 29, 1988 states at paragraph 2, "At a time prior to approximately the middle of November, 1986, Chang C. Tsuei told me a measurement he made on T<sub>c</sub> superconducting material which he said were received from Georg Bednorz and K. A. Mueller, two physicists working for IBM Corporation in Zurich, Switzerland ... Chang Tsuei said that he had measured resistivity versus temperature of these samples." This is again further evidence that the Mueller Bednorz superconducting samples were in the United States prior to the middle of November 1986."

In the Affidavit of Sung II Park, dated March 30, 1988, at paragraph 4, it is stated "the preparation in measurement of the aforementioned superconducting samples occurred at a date prior to November 15, 1986, and to the best of my recollection, occurred on or about November 9, 1986, the date when a Helium dower was pumped down preparatory to taking the actual measurement." Therefore, since measurements were taken prior to the date of publication of the Asahi Shinbum article, which was November 28, 1986 the invention was reduced to practice in the US prior to the publication date of the Asahi Shinbum article.

At page 11 of the Examiner's Action dated April 19, 1996, in the paragraph labeled i, the Examiner states "the applicants argue that Sung II Park affidavit of March 30, 1988 states at para. 4 that measurements were taken of a superconductive sample on or before November 9, 1986, to the best of affiants recollection, or no later than

November 15, 1986. The document evidence is not deemed to support that argument, however." In the paragraph marked (1) on page 11 of PA, the Examiner states "plots of those measurements are missing. See the Cheng C. Tseui affidavit of March 30, 1998, para. 6." This statement comes directly out of Cheng Tseui's Declaration.

Notwithstanding, Cheng Tseui's Declaration says the measurements were made, that the plots that were taken were missing. The last sentence of this paragraph states "I believe that they may have been inadvertently thrown away when the laboratory was subsequently extensively cleaned." The Examiner further states "a hand-drawn diagram with the indication of a vacuum pumped down on November 8, 1988 also is not deemed to show that the measurements were taken." The Examiner is referring to paragraph 5 of the Cheng Tseui Declaration and Exhibit C which contains the hand-drawn figure.

At paragraph (2) of page 11 of the Examiner's Action dated April 19, 1996, the Examiner points to cablegrams sent by Dr. Greene to applicants in Zurich which are attached as Exhibit B to his Declaration. The Examiner states "Dr. Greene reports that no indication of superconductivity has been seen in his specific heat measurements for temperature 4-35°K." The Examiner fails to note that in the same cablegram dated November 11, 1986, Dr. Greene states "this is not really too surprising given the very broad transition to have found in resistivity and susceptibility." The Examiner acknowledges that "Exhibit C has pages dated December 1, 1986 on in Exhibit D, which actually has plots and resistance versus temperature dated as early as December 3, 1986." The Examiner is conceding that high T<sub>c</sub> superconductivity was

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measured on the samples which the very same set of cablegrams and affidavit say were in the United States in the middle of November 1986. Consequently, by Examiner's own admission, samples which were in the United States were clearly shown to be superconducting as of December 3, 1986. Consequently, the samples that were in the United States as of November 9 were inherently superconducting. It is clear from the same declarations that Applicants' were communicating with Dr. Greene. It is noted that Dr. Greene's cablegram dated November 25, 1986 to applicants states he will resume work on the new superconductor and that not much will happen because of the Thanksgiving Holiday until the following week. There are cablegrams dated November 26, December 1, December 2, 1986 related to high T<sub>c</sub> superconductivity. Dr. Greene's Exhibit C has notebook pages dated December 1, 1986 to December 5, 1986. The December 5, 1986 shows T<sub>c</sub> of 26°K and 30°K. Exhibit D show a plot of R vs. T dated December 8, 1986. Clear reduction to practice is shown and clear diligence is shown from prior to the date of the Asahi Shinbum article. This was clearly done in close correspondence with the applicants. Thus, the facts clearly shown applicant's can swear behind the Asahi Shinbum reference.

The Examiner repeats the same arguments in the final rejection without rebutting Applicants' arguments and proof that their conception was in this country at their direction and that their co-inventors in this country under their direction diligently pursued a reduction to practice in this country the earliest date of which according to the Examiner is December 3, 1986.

# Applicants Invention Was Reduced To Practice In the US Before The Asahi Shinbum Article

Attachment C of this Brief is a copy of a letter from C. W. Chu to Applicants dated December 3, 1986. This letter states:

This is just to inform you that my group at the U. of Huston has reproduced your results (Z. Phys. B <u>64</u>, 189 (86)) three weeks ago. ... I believe it is superconductivity.

A journal article is an invitation to any one to reproduce the work reported therein.

Thus a reader of the article is for that purpose under the direction and control of the author. Thus Applicants invention was reduced to practice in this country prior to the date of the Asahi Shinbum article at the direction and control of Applicants.

The Examiner in the final rejection repeats the earlier rejection stating:

- i. The applicants argue that the Sung 11 Park Affidavit of March 30, 1988 states at para. 4 that measurements were taken of a superconductive sample on or before November 9, 1986, to the best of the affiant's recollection, or no later than November 15, 1986. The documentary evidence is not deemed to support that argument, however. See MPEP 715.07.
- (1) Plots of those measurements are missing. See the Chang C. Tsuei Affidavit of March 30, 1988, para. 6. A hand-drawn diagram with the

indication of vacuum pumping on November 9, 1988 also is not deemed to show that the measurements were taken.

- (2) Moreover, the other evidence in the record appears to show that high temperature superconductivity was not attained in this country as of November 9 or 15, 1986. The March 30, 1986 Declaration of Richard L. Greene includes a series of cablegrams sent by Dr. Greene to the applicants in Zurich, Switzerland as Exhibit B. On both November 11, 1986 and November 14, 1986, Dr. Greene reports that no indication of superconductivity has been seen in his specific heat measurements for temperatures of 4-35'K. Exhibit C has pages dated December 1, 1986 on, and Exhibit D, which actually has plots of resistance vs. temperature, has an earliest date of December 3, 1996.
- i. The applicants argue that "Praveen Chaudhari brought these samples back to the U.S. when he returned after visiting (the inventors) on or about October 16, 1986. When these samples came into the United States since they were inherently superconductive as claimed, the invention was essentially reduced to practice in the United States on that date." As stated before repeated above, however, the applicants were unable to show the attainment of superconductivity any earlier than December 3, 1986 in this country. Again, the present invention is directed to the method of superconducting electricity. That method apparently was not reduced to practice before December 3, 1986.
- ii. The applicants further urge that they have shown clear diligence from before November 28, 1986 until actual reduction at or around December 3, 1986. Nevertheless, the actual reduction in this country is deemed to have occurred on December 3, 1986, which is after the publication date for the reference.

- iii. The applicants assert that they should be entitled to a one-year grace period for their own published invention, but this prior art rejection is based on 35 USC 102(a) because the author of that reference is a different inventive entity.
- The applicants' proposed priority date for the EPO application is January 23, 1987, which is after the December 1986 dates show by the Richard L. Greene Affidavit.

In these repeated rejections the Examiner has not responded to Applicants' arguments and thus has not rebutted any of them. Therefore, the Examiner concedes that Applicants' conception was in this country at their direction prior to the date of the Asahi Shinburn article and that coworkers in this country at Applicants' direction diligently reduced their invention to practice in the United States. Applicants petitioned for withdrawal of the final rejection requesting the reasons for why the 1.132 affidavits submitted did not effectively swear behind the Asahi Shinburn article. The petition was denied by the Director of Group 1700 without rebutting applicants proof of conception and diligence. Therefore, the Director of Group 1700 concedes that Applicants' conception was in this country at their direction prior to the date of the Asahi Shinburn article and that coworkers in this country at Applicants' direction diligently reduced their invention to practice in the United States. This necessarily requires a finding that the Asahi Shinburn article is not a valid §102 reference.

Therefore, the Board is respectfully requested to reverse the rejection of claims 24-26, 86-90, 96-135 and 137-177 under 35 USC § 102(a) as anticipated by Asahi Shinbum article.

### **CLAIM REJECTIONS - 35 USC § 103**

Claims 24-26, 86-90, 96-135 and 137-177 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over Asahi Shinbum article.

Claim 136 has been allowed over the Asahi Shinbum article since according to the Examiner the examples in the present specification are deemed to show criticality for the formula of claim 136.

Since the present application was filed within one year of Applicants' article, Applicants' article is not a reference as to them (even though it would be a reference as to a third party). Thus the only portion of the Asahi Shinbum article relevant to Applicants' claims is:

A new ceramic with a very high  $T_c$  of 30K of the superconducting transition ... is an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties.

Since the Asahi Shinbum article refers to " $T_c$  of 30K" and since each of Applicants claims recites  $T_c > 26$   $^{\circ}$ K, the Asahi Shinbum article alone cannot anticipate  $T_c > 26$   $^{\circ}$ K since to be an anticipation a reference must contain all the limitations of the claim it is said to anticipate. Also, the Asahi Shinbum article provides no teaching of how to made the "new ceramic". A reference which does not provide a method of making a composition cannot anticipate a claim to the composition and thus to a use of that new composition. Also, the Asahi Shinbum article has no specific embodiment of the new composition. Thus it cannot anticipate under 35 USC 102. If as to Applicants the Asahi Shinbum article is not a valid .§102 reference , the Asahi Shinbum article cannot render Applicants' claims obvious since to do so would render the Asahi Shinbum article a valid .§102 against Applicants.

As state above the examiner's characterization of the Asahi Shinbum article does not accurately represent the teaching of this article.

Applicants' article which is incorporated in applicants' specification and priority document is directed to a Ba-La-Cu-O system. Applicants' article was submitted for publication on April 17, 1986 to Z. Phys. B from the IBM Zurich Research Laboratory in Switzerland. Thus the only teaching in the Asahi Shinbum article relevant to Applicants' claimed invention is the teaching in Applicants' article which is incorporated by reference in the present application and referred to in the priority document. The Asahi Shinbum article provides no teaching relevant to applicants' claimed invention other than the teaching in Applicants' article on how to make the ceramic "newly discovered" by applicants. The Asahi Shinbum article states the Applicants' "newly discovered" ceramic "is an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties". This is described in detail in Applicant's article. Thus the examiner is stating that it is obvious to a person of skill in the art to practice the invention of all of applicants non-allowed claims from the teaching of the Asahi Shinbum article which is the teaching of Applicants' specification which incorporates applicants' article by reference. Applicants note that they received the Nobel Prize in Physics in 1987 for their discovery reported in applicants article. Since Prof. Tanaka apparently only reproduced applicants work based on applicants' article, he did not share in the Nobel prize. Therefore, the examiner is stating that all of applicants non-allowed claims are obvious in view of applicants' teaching.

Alternatively, the examiner is stating that all of applicant's non-allowed claims are obvious in view of the Asahi Shinbum article's statement that "[a] new ceramic with a very high T<sub>c</sub> of 30 K of the supper conductive transition has been found. ... The ceramic newly discovered, is an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties. " Thus the examiner is stating that once a person of ordinary skill in the art knows that "an oxide compound of La and Cu with Barium which has a structure of the so-called perovskite and shows metal-like properties " has a "a very high T<sub>c</sub> of 30 K" the inventions of all of applicants non-allowed claims can be made and used by such a person of ordinary skill in the art without any additional teaching other than what is known by a person of ordinary skill.

Applicants acknowledge that if a material is known to be a superconductor, a person of skill in the art would know to cool the material to below the  $T_c$  and to flow a superconducting current therein at that temperature. Prior to the date of Applicants' invention, which is at least as early as the date on which Applicants' article was published by Z. Phys. B no one knew that transition metal oxides had a  $T_c > 26 \text{ K}$  except for Applicants. As stated above the Asahi Shinbum article describes no invention other than that of Applicants and is thus not a valid reference.

Therefore, the Board is respectfully requested to reverse the rejection of claims 24-26, 86-90, 96-135 and 137-177 under 35 USC §103 as obvious over the Asahi Shinbum article.

#### **OBJECTION TO THE SPECIFICATION UNDER 35 USC 112**

The examiner states:

5. The specification is objected to under 35 U.S.C. § 112, first paragraph, as failing to provide an enabling disclosure commensurate with the scope of the claims.

Applicants disagree. The specification incorporates by reference Applicants' article. In the rejection under 35 USC 102 and 103 over the Asahi Shinbum article, the examiner has stated that a person of skill in the art is enabled to practice the inventions of applicants' non-allowed claims by the mere statement that a Ba-La-Cu-O compound has been confirmed to be superconductive as described by Applicants in their article. The examiner's objection to the specification is inconsistent with the examiner's rejection under 35 USC 102(a) and 103(a).

The text of the Asahi Shinbum article merely states that Prof. Tanaka reproduced Applicants' work. Thus, the Examiner has stated that all of the inventions of applicants' non-allowed claims are contained within the Asahi Shinbum article and thus are fully enable by Applicants' article. The Examiner has also rejected all of applicants' non-allowed claims under 35 USC §103(a) in view of the Asahi Shinbum article. This means that nothing more is needed than what is described in Applicants' article and ordinary skill to practice all of Applicants non-allowed claims.

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#### The examiner further states:

a. The present specification is deemed to be enabled only for compositions comprising  $Ba_xLa_{5^-x}CuO_Y$ .

The examiner again used the terminology "deemed" which means that it is the examiner's "opinion". The examiner has introduced no factual evidence to support the examiner's "opinion" that "[t]he present specification is deemed to be enabled only for compositions comprising Ba<sub>x</sub>La<sub>5-x</sub>CuO<sub>Y</sub>." As shown below, the specification cites a substantially large number of specific compositions, and provides sufficient teaching to enable a person of skill in the art to practice Applicants' invention.

#### The examiner further states:

The art of high temperature (above 30 K) superconductors is an extremely unpredictable one. Small changes in composition can result in dramatic changes in or loss of superconducting properties. The amount and type of examples necessary to support broad claims increases as the predictability of the art decreases. Claims broad enough to cover a large number of compositions that do not exhibit the desired properties fail to satisfy the requirements of 35 USC 112. Merely reciting a desired result does not overcome this failure. In particular, the question arises: Will any layered perovskite material containing copper exhibit superconductivity? Also, does any stoichiometric combination of rare earth, an alkaline earth, and copper elements result in an oxide superconductor?

The examiner provides no factual evidence to support the statement "[t]he art of high temperature (above 30 K) superconductors is an extremely unpredictable one." This is an opinion of the examiner. The examiner should withdraw the rejection, provide

factual evidence to support the opinion or submit an examiner's affidavit under MPEP 706.02(a) qualifying himself as an expert in the art of high Tc superconductors to offer such a conclusory opinion. Applicants requested such an Examiner's affidavit. Since the Examiner has not provided such an affidavit, the Examiner's opinions should be disregarded. It is Applicants' teaching that controlling the amount of the constituents of the composition, such as oxygen content, effect the superconductive properties of the composition. It is a matter of routine experimentation to find the optimum constituents, such as oxygen content, for a particular high T<sub>c</sub> superconducting composition. Applicants do not have to provide experimental results for every composition that fall within the scope of their claims when a person of skill in the art exercising routine experimentation has a reasonable expectation of success following applicants teaching to achieve a composition through which can be flowed a superconducting current according to the teaching of Applicants' specification.

According to In re Angstadt 190 USPQ 214, 218 in an unpredictable art, §112 does not require disclosure of a test with every species covered by a claim. The CCPA states:

To require such a complete disclosure would apparently necessitate a patent application or applications with "thousands" of examples or the disclosure of "thousands" of catalysts along with information as to whether each exhibits catalytic behavior resulting in the production of hydroperoxides. More importantly, such a requirement would force an inventor seeking adequate patent protection to carry out a prohibitive number of actual experiments. This would tend to discourage inventors

from filing patent applications in an unpredictable area since the patent claims would have to be limited to those embodiments which are expressly disclosed. A potential infringer could readily avoid "literal" infringement of such claims by merely finding another analogous catalyst complex which could be used in "forming hydroperoxides."

The Examiner provides no evidence to support he examiner's statement that "[t]he amount and type of examples necessary to support broad claims increases as the predictability of the art decreases." The examiner has provided no evidence that the predictability of art of high Tc superconductivity is low. The Examiner's statement that "[c]laims broad enough to cover a large number of compositions that do not exhibit the desired properties fail to satisfy the requirements of 35 USC 112." implies that applicants claims "cover a large number of compositions that do not exhibit the desired properties" of high T<sub>c</sub> superconductors. The Examiner has provided no evidence to support the examiners' implication. In fact, the claims do not cover any compositions that do not exhibit the desired properties of high T<sub>c</sub> superconductors. Applicants claims only cover superconductors having T<sub>c</sub> > 26°K which carry a superconductive current. Applicants' claims are not composition of matter claims.

The Board's attention is directed to the following comments from the specification.

The specification states at page 1, lines 5-10, that "This invention relates to ... superconducting compositions including copper and/or transition metals."

The specification further states at page 5, lines 2-9 that:

"It is another object of the present invention to provide novel superconductive materials that are multi-valent oxides including transition metals, the compositions having a perovskite-like structure."

It is a further object of the present invention to provide novel superconductive compositions that are oxides including rare earth and/or rare earth-like atoms, together with copper or other transition metals that can exhibit mixed valent behavior."

The specification further states at page 8, lines 1-11, that "[A]n example of a superconductive composition having high T<sub>c</sub> is the composition represented by the formula RE-TM-O, where RE is a rare earth or rare earth-like element, TM is a nonmagnetic transition metal, and O is oxygen. Examples of transition metal elements include Cu, Ni, Cr etc. In particular, transition metals that can exhibit multi-valent states are very suitable. The rare earth elements are typically elements 58-71 of the periodic table, including Ce, Nd, etc. If an alkaline earth element (AE) were also present, the composition would be represented by the general formula RE-AE-TM-O."

And at page 7, lines 14-15, the specification states that "the rare earths site can also include alkaline earth elements."

The specification further states at page 11, lines 19-24, that "An example of a superconductive compound having a layer-type structure in accordance with the present invention is an oxide of the general composition RE<sub>2</sub>TMO<sub>4</sub>, where RE stands for the rare earths (lanthanides) or rare earth-like elements and TM stands for a transition metal."

The composition RE<sub>2</sub>TMO<sub>4</sub>:RE is referred to at page 24, lines 5-9; RE<sub>2-x</sub>TM<sub>x</sub>O<sub>4-y</sub> is referred to at page 25, lines 19-21.

The following specific compounds are recited in the application:

Ba<sub>4</sub>La<sub>5-x</sub>Cu<sub>5</sub>O<sub>5(3-y)</sub> at page 10, lines 4, 10, 14.

La<sub>2-x</sub>Ba<sub>x</sub>CuO<sub>4-y</sub> at page 12, line 13

La<sub>2-x</sub>Ba<sub>x</sub>NiO<sub>4-y</sub> at page 12, line 13

La<sub>2-x</sub>Sn<sub>x</sub>NiO<sub>4-y</sub> at page 12, line 17

Ce<sub>2-x</sub>Cu<sub>x</sub>NiO<sub>4-y</sub> at page 12, line 19

La₂CuO₄ at page 12, line 21

 $La_2CuO_{4-y}$  with  $Sr^{2x}$ ,  $Ba^{2x}$  and  $Ca^{2x}$  substitution at page 13, line 17

La<sub>2-x</sub>Sn<sub>x</sub>CuO<sub>4-y</sub> at page 17, line 21

La<sub>2-x</sub>Ca<sub>x</sub>CuO<sub>4-y</sub> at page 17, line 21

La<sub>2-x</sub>Ba<sub>x</sub>CuO<sub>4-y</sub> at page 18, line 6

La<sub>2</sub>CuO<sub>4</sub> :Ba at page 18, line 15

La<sub>2</sub>CuO<sub>4</sub>:Ba at page 24, line 6

Nd<sub>2</sub>NiO<sub>4</sub>: Sn at page 24, line 9

La<sub>2</sub>CuO<sub>4-y</sub> doped with Sn<sup>2x</sup>, Ca<sup>2x</sup> and Ba<sup>2x</sup> at page 25, lines 6-18

Other compounds are given in the articles to B. Raveau, in Mat. Res. Bull., Vol. 20 (1985) pp. 667-671, and to C. Michel et al. in Rev. Claim. Min. 21 (1984) 407, both of which are incorporated by reference at page 13, lines 4-5.

The Examiner cites In re Fisher, 166 USPQ 18, In re Angstadt and Griffen, 150 USPO 214, and In re Golianni, 195 USPQ 150, in support of the statement "[t]he amount and type of examples necessary to support broad claims increases as the predictability of the art decreases".

The claims under appeal In re Fisher are directed to increasing the potency of substances containing ACTH hormones for injection into human beings. In regards to the rejection for insufficient disclosure under 35 USC 112 the CCPA states that:

"the issue thus presented is whether an inventor with the first to achieve potency of greater that 1.0 for certain types of compositions, which potency was long designed because of its beneficial effects on humans, should be allowed to dominate *all* compositions having potencies greater 1.0, thus including future compositions having potencies in excess of those obtainable from his teachings plus ordinary skill." 166 USPQ 18, 23-24 (emphasis in the original).

The CCPA goes on to say in In re Fisher that:

"It is apparent that such an inventor should be allowed to dominate the future patentable inventions of others where those inventions were based in some way on his teachings. Such improvements, while unobvious from his teachings, are still within his contribution, since the improvement was made possible by his work. It is equally apparent, however, that he must not be committed to achieve this dominance by claims which are insufficiently supported and hence, not in compliance with the first paragraph of 35 USC 112. That paragraph requires that the scope of the claims must bear a reasonable correlation to the scope of enablement provided by the specification to persons of ordinary skills in the art... In cases involving unpredictable factors, such as most chemical reactions... the scope of enablement obviously varies inversely with the degree of unpredictability of the factors involved." (166 USPQ 18, 24)

Applicants of the present invention have provided the first teaching that transition metal oxides can form a superconductor having a critical temperature in excess of 26°K, therefore, "is apparent that such an [applicant] should be allowed to dominate the future patentable inventions of others when those inventions [are based in some way on applicants] teaching" as stated by the CCPA in In re Fisher Supra.

In the present invention Applicants are acknowledged to be the pioneers of high T<sub>c</sub> superconducting metal oxides. The Examiner has produced no evidence that invention which come within the scope of Applicants' claim cannot be achieved by persons of skill in the art based on Applicants teaching. The affidavits of Mitzi, Dinger, Tsuei, Shaw and Duncombe and the book of Poole et al. state it is straight forward to use the general principles of ceramic science to make high T<sub>c</sub> transition metal oxide superconductors following Applicants teaching.

The claimed invention in re Angstadt and Griffen (190 USPQ 214) involves a methods of catalycally oxidizing alkylaromatic hydrocarbons to form a reaction comprising the corresponding hydroperoxides. The method employs catalysts. The Examiner rejected all the claims under 35 USC 112, first and second paragraphs. The Board's rational for affirming the Examiner's rejection was directed primarily to the enablement required of the first paragraph.

The CCPA reversing stated that:

"what is a maximum concern in the analysis of whether a particular claim is supported by the disclosure in an application, is whether the disclosure contains sufficient teaching regarding the subject matter of the claims as enabled one of skill in the art to make and to use the claimed invention. These two requirements 'how to make' and 'how to use' have some times been referred to in combination as the 'enablement requirement'... The relevancy may be summed up as being whether the scope of enablement provided to one of ordinary skill in the art by the disclosure as such as to be commensurate with the scope or protection sought by the claims. (190 USPQ 214,47 citing In re Moore 169 USPQ).

In the attached affidavits under 37 CFR 132, Dr. T. Dinger, Dr. Tsuei, Dr. Shaw, Mr. Duncombe and Dr. D. Mitzi state:

"That once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the above-identified patent application, which includes all known principles of ceramic fabrication, can make the transition metal oxide compositions encompassed by claims 24-26, 86-90 and

96-108, without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art. This is why the work of Bednorz and Müller was reproduced so quickly after their discovery and why so much additional work was done in this field within a short period of their discovery."

In the paragraph at the bottom of page 15 of the specification, it is stated that: in regard to compositions according to the present invention that "their manufacture generally follows the known principles of ceramic fabrication." Thereafter, an example of a typical manufacturing process is given.

The CCPA in In re Angstadt and Griffen further states that:

"we cannot agree with the Board that Appellants' disclosure is not sufficient to enable one of ordinary skill in the art to practice the invention without undue experimentation. We note that many chemical processes and catalytic processes particularly, are unpredictable, ..., and the scope of enablement varies inversely with the degree of unpredictability involved... The question, then, whether in an unpredictable art, section 112 requires the disclosure of a test with every species covered by a claim. To require such a complete disclosure will apparently necessitate a patent application or applications with 'thousands' of examples... . More

importantly, such a requirement would force an inventor to seek adequate patent protection to carry out a prohibited number of natural experiments. This would tend to discourage inventors in filing patent applications in an unpredictable area since the patent claim would have to be limited those embodiments which are expressly disclosed. A potential infringer could readily avoid 'infringement of such claims' by merely finding another analogous (example) which could be used..." 190 USPQ 124, 218.

The CCPA in In re Angstadt further goes on to say

"having decided that appellants are *not* required to disclose every *species* encompassed by the claims even in an unpredictable art such as the present record presents, each case must be determined on its own facts." 190 USPQ 214, 218. (emphasis in the original).

In regards to the catalyst In re Angstadt and Griffen CCPA further states:

"since appellants have supplied the list of catalysts and have taught how to make or how to use them, we believe that the experimentation required to determine which catalyst will produce hydroperoxide would not be undo and certainly would not 'require ingenuity beyond that to be expected of one of ordinary skill in the art'. 190 USPQ, 214, 218 in re Field v. Connover 170 USPQ, 276, 279 (1971).

As stated in the affidavits of Dr. Dinger, Dr. Tsuei, Dr. Shaw, Mr. Duncombe and Dr. Mitzi, to make the high temperature superconductors encompassed by Applicants' claims, using the teaching of the present invention would not require ingenuity beyond that expected of one of ordinary skill in the art.

The CCPA in In re Angstadt further states that:

"the basic policy of the Patent Act, which is to encourage disclosure of inventions and thereby to promote progress in the useful arts. To require disclosures in patent applications to transcend the level of knowledge of those skilled in the art would stifle the disclosure of inventions in fields man understands imperfectly." 190 USPQ 214, 219.

The CCPA further states that:

"the certainty which the law requires in patents is not greater than is reasonable." 242 USPQ, 270-271, cited in In re Angstadt. 190 USPQ 214, 219.

In re Angstadt further states at 190 USPQ 219:

We note that the PTO has the burden of giving reasons, supported by the record as a whole, why the specification is not enabling. In re Armbruster, 512 F.2d 676, 185 USPQ 152 (CCPA 1975). Showing that the disclosure entails undue experimentation is part of the PTO's initial burden under Armbruster; this court has never held that evidence is necessary for any experimentation, however slight, is sufficient to require the applicant to prove that the type and amount of experimentation needed is not undue.

By calling the claimed "invention" the "scope of protection sought" the dissent obscures the problem and frustrates the intended operation of the patent system. Depriving inventors of claims which adequately protect them and limiting them to claims which practically invite appropriation of the invention which avoiding infringement inevitably has the effect of suppressing disclosure. What the dissent seem to be obsessed with is the thought of catalysts which won't work to produce the intended result. Applicants have enabled those in the art to see that this is a real possibility, which is commendable frankness in a disclosure. Without undue experimentation or effort or expense the combinations which do not work will readily be discovered and, of course, nobody will use them and the claims do not cover them. The dissent wants appel lants to make everything predictable in advance, which is impracticable and unreasonable.

We hold that the evidence as a whole, including the inoperative as well as the operative examples, negates the PTO position that persons of ordinary skill in this art, given its unpredictability, must engage in undue experimentation to determine which complexes work. The key word is "undue," not "experimentation."

The only facts which the Examiner offers as evidence of unpredictability are examples provided in Applicants' specification. The CCPA in In re Angstadt says that

this is "commendable frankness" which is not to be held against Applicants. The Examiner has provided no evidence that a person of skill in the art has to engage in undue experimentation to practice Applicants non-allowed claims. The affidavits of Mitzi, Dinger, Tsuei, Shaw and Duncombe and the book of Poole et al. indicate that persons of skill in the art do not have to engage in undue experimentation to practice Applicants' invention.

The Examiner cited In re Colianni 195 USPQ 150 which applicants believe is not on point since in In re Colianni "[t]here is not a single specific example or embodiment by way of an illustration of how the claimed method is to be practiced." (195 USPQ 150, 152). In contradistinction as noted above, there are numerous examples cited in applicants' specification and incorporated references. Thus this decision is not on point.

"Showing that the disclosure entails undue experimentation is part of the PTO's initial burden." In re Armbruster 185 USPQ 152, 504.

"The practical approach followed consistently by [the CCPA] ..., places the initial burden on the PTO to show that the enabling disclosure is not commensurate in scope with the claim. Upon such a showing, the burden of rebuttal shifts to applicants". In re Coliani 195 USPQ 150.

"However, [the CCPA] has made it clear that the Patent and Trademark Office must substantiate its rejections for lack of enablement with reasons" In re Armbruster 185 USPQ 152, 153.

The Examiner has merely asserted without support that "the art of high temperature superconductivity is unpredictable...".

The CCPA in In re Marzocchi, 58 CCPA 1069, 439 F. 2d 220, 169 USPQ 367, 369-370 (1971) states:

"The only relevant concern of the Patent Office under these circumstances should be over the *truth* of any such assertion. The first paragraph of §112 requires nothing more than objective enablement. How such a teaching is set forth, either by the use of illustrative examples or by broad terminology, is of no importance.

As a matter of Patent Office practice, then, a specification disclosure which contains a teaching of the manner and process of making and using the invention in terms which correspond in scope to those used in describing and defining the subject matter sought to be patented *must* be taken as in compliance with the enabling requirement of the first paragraph of §112 *unless* there is reason to doubt the objective truth of the statements contained therein which must be relied on for enabling

support. Assuming that sufficient reason for such doubt does exist, a rejection for failure to teach how to make and/or use will be proper on that basis; such a rejection can be overcome by suitable proofs indicating that the teaching contained in the specification is truly enabling...

[I]t is incumbent upon the Patent Office, whenever a rejection on this basis is made, to explain *why* it doubts the truth or accuracy of any statement in a supporting disclosure and to back up assertions of its own with acceptable evidence or reasoning which is inconsistent with the contested statement. Otherwise, there would be no need for the applicant to go to the trouble and expense of supporting his presumptively accurate disclosure. [Emphasis in original footnote deleted].

Applicants have submitted herewith affidavits of Dr. Mitzi, Dr. Tsuei, Dr. Shaw, Mr. Duncombe and Dr. Dinger under 37 CFR 132 which state, as quoted above, that once a person of skill in the art knows of applicants' work, the compositions encompassed by the claims under experimentation, can be made using the teaching of applicants without undue experimentation.

Thereby rebutting the Examiner's statement that:

"[the specification ... [fails] to provide an enabling disclosure commensurate with the scope of the claims."

The Examiner cites In re Cooks and Cosden Oil v. American Hoechst to support the statement "[c]laims broad enough to cover a large number of compositions that do not exhibit the desired properties fail to satisfy the requirement of 35 USC 112." Applicants claims do not read on any inoperative specifies since Applicants' claims are method of use claims. A composition which does not have a  $T_c > 26$ °K is not within the scope of the claims. Thus these decisions are not on point.

The Examiner cites In re Corkill as support for this statement "[m]erely reciting a desired result does not overcome this failure". The CAFC held "[c]laims which include a substantial measure of inoperatives ... are fairly rejected under 35 USC 112."

Applicants' claims include no inoperatives. Since Applicants claims are method of use claims they are functional and thus exclude inoperatives. "[T]he use of functional language is sanctioned specifically by ... section 112." In re Angstadt 190 USPQ 214, 217.

The Examiner cited Brenner v. Manson for the statement "a patent is not a hunting license. It is not a reward for the search, but a reward for its successful conclusion." The claim in question was a method of making a composition. The composition had no known use. The method was found to lack utility and thus is not be patentable. This is not relevant to §112, first paragraph. Thus this decision is not on point. Moreover, Applicants have had a successful conclusion, they won a Nobell Prize and initiated and enabled the high T<sub>c</sub> transition metal oxide art.

The Examiner queries "[w]ill any layered perovskite material containing copper exhibit superconductivity?" and "does any stoichiometric combination of rare earth, an alkaline earth, and copper elements result in an oxide superconductor?" Since applicants claims are directed to methods of using oxide compositions, applicants' claims read on only those layered perovskite materials which exhibit superconductivity with a  $T_c > 26~^{\circ}$ K and do not read on methods of use of oxide compositions which are not superconductive. Thus the Examiner's queries is not relevant to applicants claims. Applicants are not claiming a composition which is a high  $T_c$  superconductor. Thus applicants claims do not read on any layer perovskite, or any other stoichiometric combination, but only those carrying a high  $T_c$  superconducting current. Method of use claims are inherently narrower in scope than composition claims.

The paragraph bridging pages 13 and 14 refer to Ba-La-Cu-O systems having different crystallographic phases having Cu<sup>3+</sup> and Cu<sup>2+</sup> ions or Ni<sup>3+</sup> and Ni<sub>2+</sub> ions.

Claim 103 of the present invention recites "a copper oxide compound having a layer-type-perovskite-like crystal structure, the copper oxide compound including at least one rare-earth or rare-earth-like element, and at least one alkaline-earth element". In regard to the stated elements, the rare earth elements are defined in the specification at pate 7, lines 9-12 to the "a group IIIB element, such as La." Group IIIB includes Sc, Y, La and Ac, rare earth-like or near rare earth. The rare earth elements are elements 58 to 71. This group contains four elements from group IIIB and fourteen

elements from the rare-earth for a total of 18 elements. The alkaline earths contain the elements of Group A which has 6 elements.

The Examiner further states:

It should be noted that at the time the invention was made, the theoretical mechanism of superconductivity in these materials was not well understood. That mechanism still is not understood. Accordingly, there appears to be little factual or theoretical basis for extending the scope of the claims much beyond the proportions and materials actually demonstrated to exhibit high temperature superconductivity. A "patent is not a hunting license. It is not a reward for the search, but a reward for its successful conclusion".'

The Examiner has provided no evidence to support the statement "that at the time the invention was made, the theoretical mechanism of superconductivity in these materials was not well understood. That mechanism is still not understood." Applicants request the Examiner to introduce evidence to support this statement or to place an Examiner's affidavit under MPEP 706.02(a) qualifying the Examiner as an expert to make this statement. The Examiner further states "there appears to be little factual or theoretical basis for extending the scope of the claims much beyond the proportions and materials actually demonstrated to exhibit high temperature superconductivity." This is the Examiner's unsupported opinion. The five affidavits of Mitzi, Dinger, Tsuei,

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Shaw and Duncombe under 37 CFR 1.132 and the book to Poole et al. described below provide factual evidence supporting Applicants position that once a person of skill in the art knew from Applicant's article that transition metal oxides were high Tc superconductors, it was a matter of routine application of the general principles of ceramic science to fabricate transition metal oxide superconductors other than those actually made by applicants. Quoting "Brenner v. Manson", 283 US 518, 148 USPQ 689, the Examiner further states that a "patent is not a hunting license. It is not a reward for the search, but a reward for its successful conclusion." The evidence introduced by Applicants clearly shows that Applicant's article upon which the present application is based had a very successful conclusion. Applicants started the field of high Tc superconductivity. All the further developments were based on Applicants teaching. Moreover, the issue in Brenner v. Manson was the patentability of a method to fabricate a composition. The composition had no use. The method was found not patentable for lack of utility. To issue a patent for such a process would be granting a hunting license for a utility that may occur in the future. This case has noting to do with §112 enablement.

In view of the above Applicants request the Board to reverse the Examiner's objection to the specification.

## CLAIM REJECTIONS UNDER 35 USC §112, FIRST PARAGRAPH

Claims 24-26, 86-90, 96-113, 129-131, 134, 135, and 139-177 are rejected under 35 U.S.C. § 112, first paragraph, for the same reasons set forth by the Examiner in the objection to the specification. Applicants disagree for the same reasons why Applicants disagreed to the objection to the specification above. The Examiner provides specific comments only in regards to claims 24, 86, 88, 96 and 103.

Applicants disagree with the Examiner's statement that Applicants' arguments filed April 11, 1996, January 3, 1996, and September 29, 1995, paper nos. 53, 50, and 51, as well as the Affidavits filed September 29, 1995 and January 3, 1996, paper nos. 49 and 52, have been fully considered but they are not deemed to be persuasive. The arguments of those papers are incorporated herein by reference.

The Examiner further states:

"The additional case law and arguments by the applicants have been duly noted. For the reasons that follow, however, the record as a whole is deemed to support the initial determination that the originally filed disclosure would not have enabled one skilled in the art to make and use the invention to the scope that it is presently claimed."

The Examiner again uses the word "deemed", that is, it is the Examiner's opinion unsupported by any factual evidence. The quoted passage is completely contrary to the Examiner's rejection under 35 USC 102(a) and 103(a). Under these rejections the Examiner found the Asahi Shinbum article would have enabled one skilled in the art to make and use the invention to the scope that it is presently claimed. As noted above, the Asahi Shinbum article relies upon Applicants article. Applicants' view is further supported by the five affidavits of Mitzi, Tsuei, Dinger, Shaw and Duncombe under 37 CFR 1.132 and the book of Poole which will be described below and which states that once it was known from Applicants article that transition metal oxides were superconductive at temperatures above 26°K, other high Tc transition metal oxides could be made by a person of skill in the art using the teaching of Applicants and the general teachings of ceramic science.

### The Examiner further states:

ii. The applicants quote several passages from their specification at pp. 13-15 of their September 29, 1995 Amendment, but the issue is the scope of enablement, not support. The present disclosure may or may not provide support for particular embodiments, but the issue here is the scope to which that disclosure would have taught one skilled in the art how to make and use a composition which shows the onset of superconductivity at above 26°K.

Applicants are not required to provide experimental evidence for every species within the scope of their claims. The entire filed of high T<sub>c</sub> superconductivity is based on Applicants' work, in particular, on Applicants' article. This affidavit of James W. Leonard under 37 CFR 1.132 submitted on December 15, 1998 states that 5,689 articles cited Applicants' article (Z. FurPhys. B, 64, pp. 185-193 (September 1986). This is clear evidence that Applicant article originated and enabled the filed of high T<sub>c</sub> superconductivity. It is clear by the facts presented by Applicants that they "have taught one of skill in the art how to make and use a composition which shows the onset of superconductivity at above 26°K". The Examiner has introduced no factual evidence to the contrary. The Examiner merely "deems" that Applicants' claims are not enabled.

The Examiner further states:

iii. Construed in light of that issue, the invention is not deemed to have been fully enabled by the disclosure to the extent presently claimed.

The Examiner again "deems" Applicants claims not enabled without factual support.

The Examiner further states:

(1) In their September 29, 1995 Amendment, the applicants argue that their disclosure refers to "the composition represented by the formula RE-TM-0, where RE is a rare earth or rare earth-like element, TM is a

nonmagnetic transition metal, and 0 is oxygen", and list several species such as "La<sub>2-x</sub>Ba<sub>x</sub>CuO<sub>4-y</sub>" which they indicate are found in the present disclosure.

Applicants disagree. The passages quoted in the September 29, 1995 amendment lists numerous specific compositions and generic formula as indicated above. The listed species were preferred embodiments at the time of filing the present application. Preferred embodiments do not provide a limitation to the broad scope of applicants teaching.

The Examiner further states:

(2) Notwithstanding that argument, it still does not follow that the invention is fully enabled for the scope presently claimed. The claims include formulae which are much broader than the RE-TM-0 formula cited in the disclosure. Claim 24 recites "a transition metal oxide", claim 88 "a composition", and claim 96 "a copper-oxide compound".

As noted above the specification at page 1, line 5-10, states "[t]his invention relates to ... superconducting compositions including copper and/or transition metals."

And at page 5, lines 5-9 "[i]t is [an] ... object of the present invention ... to provide novel superconductive compositions that are oxides."

The Examiner further states:

(a) The present specification actually shows that known forms of "a transition metal oxide", "a composition", and "a copper-oxide compound" do not show the onset of superconductivity at above 26 °K. At p. 3, line 20, through p. 4, line 9, of their disclosure, the applicants state that the prior art includes a "Li-Ti-O system with superconducting onsets as high as 13.7°K. " Official Notice is taken of the well-known fact that Ti is a transition metal. That disclosure also refers to "a second, non-conducting CuO phase" at p. 14, line 18.

Applicants claims are directed to "transition metal oxides", "a composition" and "a copper-oxide compound" having a  $T_c$  in excess of 26°K which is carrying a superconducting current. Applicants claims do not include in the claimed method compositions having  $T_c < 26$ °K. Thus the examples on page 3, line 20 - page 4, line 9, are not included in applicants claims. That these are transition metal oxides having  $T_c < 26$ °K does not mean that Applicants' claims directed to transition metal oxides, compositions and copper oxides having  $T_c > 26$ °K are not enabled. Applicants provide the teaching on how to fabricate such oxides having  $T_c > 26$ °K. The "second non-conducting CuO phase" referred to at page 14, line 18, again does not mean that applicants claims are not enabled. Applicants statements at page 14 is part of applicants teaching on how to achieve an oxide having a  $T_c > 26$ °K. The Examiner is attempting to use Applicants complete description of their teaching to show lack of

enablement when, in fact, this complete teaching provides full enablement by showing how samples are and are not to be prepared. Applicants have claimed their invention functionally, that is, as a method of use so the Applicants' claim do not read on inoperable species. What the Examiner "seems to be obsessed with is the thought of [transition metal oxides] which won't work to produce the intended result. Applicants have enabled those of skill in the art to see that this is a real possibility which is commendable frankness in a disclosure." In re Angstadt, Supra.

The Examiner further states:

(b) Accordingly, the present disclosure is not deemed to have been fully enabling with respect to the "transition metal oxide" of claim 24, the "composition" of claim 88, or the "copper-oxide compound" of claim 96.

Again without facts the Examiner "deems" Applicants' claims not enabled and for the reasons given above applicants disagree. The only attempt at a factual support for the Examiner's statement are the examples provided by Applicants which show  $T_c$  < 26°K. Applicants provide this teaching so that a person of skill in the art will be fully informed on how to practice Applicants invention.

The Examiner further states:

- (3) The examples at p. 18, lines 1-20, of the present specification further substantiates the finding that the invention is not fully enabled for the scope presently claimed.
- (a) With a 1:1 ratio of (Ba, La) to Cu and an x value of 0.02, the La-Ba-Cu-0 form (i.e., "RE-AE-TM-0", per p. 8, line I 1) shows "no superconductivity".

This part of Applicants' teaching providing a complete teaching which permits a person of skill in the art to be fully enabled to practice Applicants' claimed invention. The issue according to In re Angstadt (referred to above) is whether Applicants have enabled those of skill in the art to practice Applicants invention without undue experimentation. That a person of skill in the art has to do experimentation to determine suitable combinations of elements to form a composition having T  $_{\rm c} > 26^{\rm o}{\rm K}$  through which a superconductive current flows is not an indication of a lack of enablement. Thus Applicants cited examples of compositions having T  $_{\rm c} < 26^{\rm o}{\rm K}$  does not show lack of enablement.

The Examiner further states in regards to the examples at p. 18, lines 1-20:

(b) With a 2:1 ratio of (Ba, La) to Cu and an x value of 0.15, the La-Ba-Cu-0 form shows an onset of superconductivity at "Tc = 26°K". It should be noted, however, that all of the claims in this application require the critical temperature (Tc) to be "in excess of 26°K" or "greater than 26'K".

Applicants do not believe that an on set of a  $T_c$  at 26°K is different than and greater than 26°K. Greater than 26°K can mean  $T_c$  = 26°K plus some infinitesimal temperature.

The Examiner further states:

(c) Consequently, the present disclosure is not deemed to adequately enable the full scope of the present claims. Independent claims 86 and 103 may require the presence of rare earth, alkaline earth, and transition metals, but the aforementioned examples show that superconductivity is still very unpredictable. Those claims cannot be deemed to be fully enabled.

The Examiner again "deems" claims 86 and 103 as not fully enabled. This again is the Examiner's opinion. Claims 86 and 103 do not read on the examples cited in

Applicants' specification which are superconductors having  $T_c$  < 26°K. Thus these examples are not evidence that claims 86 and 103 are not enabled. The examples are part of the teaching enabling a person of skill in the art to practice the invention of claims 86 and 103.

The Examiner further states:

iv. The applicants also have submitted [five] affidavits attesting to the applicants' status as the discoverers of materials that superconduct > 26°K. Each of the affidavits further states that "all the high temperature superconductors which have been developed based on the work of Bednorz and Muller behave in a similar manner (way)". Each of the affidavits add "(t)hat once a person of skill in the art knows of a specific transition metal oxide composition which is superconducting above 26°K, such a person of skill in the art, using the techniques described in the (present) application, which includes all known principles of ceramic fabrication, can make the transition metal oxide compositions encompassed by (the present) claims ... without undue experimentation or without requiring ingenuity beyond that expected of a person of skill in the art."

Applicants acknowledge that all five affiants are the employees of the assignee of the present application. (The affidavit of Shaw and Duncombe were submitted after

the date of the final rejection and were considered as indicated by the Advisory Action dated February 25, 1999).

The Examiner states in response to the affidavits of Mitzi, Tsuei and Dinger:

(1) Those affidavits do not set forth particular facts to support the conclusions that all superconductors based on the applicants' work behave in the same way and that one skilled in the art can make those superconductors without undue experimentation. Conclusory statements in an affidavit or specification do not provide the factual evidence needed for patentability.

The Examiner cited In re Lindner, 173 USPQ 356, 358 (CCPA 1972) in support of this statement. In In re Lindner the patent applicant submitted Rule 132 affidavit based on one example to show unexpected results for a claim of broader scope. The CCPA held that "[i]t is well established that objective evidence of non-obviousness must be commensurate in scope with the claims." In re Lindner is not on point since it does not deal with the issue of enablement. A single example can enable a broader scope claim where nothing mere is needed than what is taught by Applicants or what is taught by Applicants together with what is know by a person of skill in the art.

The affidavits of Mitzi, Tsuei, Dinger, Shaw and Duncombe are statements of experts in the ceramic arts. The Examiner disagrees with these experts. But the

Examiner has not submitted an Examiner's affidavit qualifying himself as an expert to rebut the statements of Applicants affidavits.

The Examiner further states:

(2) Those affidavits do not overcome the non-enablement rejection. The present specification discloses on its face that only certain oxide compositions of rare earth, alkaline earth, and transition metals made according to certain steps will superconduct at >26°K.

Applicants disagree. The affidavits of Shaw and Duncombe cites numerous books and articles which provide the general teaching of ceramic science at the time of and prior to the filing date of the present application. The affidavit of Duncombe also provides several hundred pages copied from Mr. Duncombe's notebooks starting from before Applicants' filing date. In regards to these pages, Mr. Duncombe states "I have recorded research notes relating to superconductor oxide (perovskite) compounds in technical notebook IV with entries from November 12, 1987 to June 14, 1998 and in technical notebook V with entries continuing from June 7, 1988 to May 1989." Mr. Duncombe's affidavit list some of the compounds prepared using the general principles of ceramic science: Y<sub>1</sub> Ba<sub>2</sub> Cu<sub>3</sub> O<sub>x</sub>,

 $Y_1 \ Ba_2 \ Cu_3 \ O_3, \ Bi_{2.15} \ Sr_{1.98} \ Ca_{1.7} \ Cu_2 \ O_{8+8}, \ Ca_{(2-x)} \ Sr_x \ Cu \ O_x \ and \ Bi_2 \ Sr_2 \ Cu \ O_x.$ 

The Examiner further states:

(3) Those affidavits are not deemed to shed light on the state of the art and enablement at the time the invention was made. One may know now of a material that superconducts at more than 26 °K, but the affidavits do not establish the existence of that knowledge on the filing date for the present application. Even if the present application "includes all known principles of ceramic fabrication", those affidavits do not establish the level of skill in the ceramic art as of the filing date of that application.

The Examiner states that "these affidavits are not **deemed** to shed light on the state of the art and enablement at the time the invention was made," that is, it is the Examiner's opinion. Applicants disagree. The affidavits clearly state that all that is needed is Applicants' teaching and the ordinary skill of the art to practice applicants claimed invention. Also, 35 USC §112, does not require that enablement be determined "at the time the invention was made". This language appers in 35 USC §103, but not in 35 USC §112. Thus it is clear that it was not the intent of Congress to determine enablement at the time the invention was made in the manner sug gested by the Examiner. All that is necessary is "[t]he specification shall contain a written description ... to enable any person skilled in the art ... to make and use the same." Applicants initiated the filed of high T<sub>o</sub> superconductors. If a person of skill in the art from the description in Applicants' specification can practice applicants' claimed invention, it is enabled. Applicants' are not required to show that a person of skill in the

art had the knowledge prior to Applicants' invention. If this were the case Applicants would not be the first, sole and only inventors, since the invention would be known by others. Applicants teach ceramic processing methods to fabricate high T<sub>o</sub> superconductors. This uses general principles of ceramic science known prior to the filing date of the present application. Thus applicants' claims are fully enabled. The Examiner has provided no evidence to the contrary. The Examiner has produced no evidence to demonstrate that a person of skill in the art, at the time of applicants filing date, could not practice the claimed invention from Applicants teaching. The utilization of such teaching to practice Applicants' claimed invention was not known prior to Applicants' filing date. That is Applicants' discovery and thus why they are entitled to their claimed invention.

The Examiner further states:

(4) It is fully understood that the applicants are the pioneers in high temperature metal oxide superconductivity. The finding remains, nonetheless, that the disclosure, is not fully enabling for the scope of the present claims.

If Applicants pioneered the field of high T<sub>c</sub> superconductivity, that is, they initiated the substantial worldwide effort to validate their discovery and to synthesize others specific embodiment of their generic and specific teaching, then applicants should be entitled to generic claims since other based their work on applicants' teaching.

The Examiner has cited the following seven decisions, which have been discussed in detail above, in support for the determination of non-enablement: <u>In re Fisher</u>, 166 USPQ 18, 24; and <u>In re Angstadt and Griffen</u>, 190 USPQ 214, 218. <u>In re Colianni</u>, 195 USPQ 150, 153, 154 (CCPA 1977). <u>In re Cook</u>, 169 USPQ 298, 302; and <u>Cosden Oil v. American Hoechst</u>, 214 USPQ 244, 262. <u>In re Corkill</u>, 226 USPQ 105, 1009. <u>Brenner v. Manson</u>, 383 US 519, 148 USPQ 689.

The Examiner has not applied the rational of these decisions. In fact, in the prosecution Applicant pointed out that the Examiner seems to have specifically avoided applying this case law and, consequently, applicants take the Examiner's silence as concurrence in the manner that applicants have applied this case law. In response to this the Examiner states "[n]ot withstanding the applicants' commentary on caselaw, the April 15, 1997 Office Action, paper no. 54, sets forth the factual basis for the determination of non-enablement at pp. 5-10." It is applicants view that the Examiner is misapplying this case law.

In this regard the Examiner further states:

(3) The applicants still further argues "that the Examiner does not rebut the case law and argument provided by applicants on (pages) 15-25 of their September 29, 1995 amendment which addresses (these issues) in detail." The point remains, nevertheless, that there appears to be a

concurrence as to the applicable caselaw. That caselaw speaks for itself What has been fully addressed in the previous Office Action and repeated above is the factual basis for the determination of non-enablement for the scope of the present invention.

Case law does not speak for itself. The rule of a case depends on the facts of the case. The fact circumstances in the case must be compared with those of the present application to justify application of the decision of the case. The Examiner has not done this. Applicants have shown above that the case law when properly applied support Applicants' position that their claims are enabled and does not support the Examiner's position that the non-allowed claims are not enabled.

#### The Examiner states:

ii. The applicants urge that "their disclosure supports a substantially broader scope than (particular) species." With respect to transition metals, the applicants point to the support in their disclosure and argue that they were enabled for transition metals other than just copper. Again, however, it is noted that high temperature superconductivity is a highly unpredictable art. In view of the record as a whole, it is again determined that one skilled in the art would not have been enabled to practice the presently disclosed invention with transition metals other than copper.

"The first paragraph of §112 requires nothing more than objective enablement , how such teaching is set forth , either by the use of illustrative examples or by broad terminology is of no importance" In re Marzochi, Supra. The Examiner has provided no factual evidence to support the Examiner's statement that the field of high T  $_{\rm c}$  superconductivity is unpredictable. The Examiner has attempted to support this position based on Applicants' teaching. That teaching is part of Applicants' enabling disclosure. In the last sentence of the passage above the Examiner implies that applicants claims directed to copper are enabled. Applicants teach transition metals. Copper is a transition metal. Applicants teaching enable persons of skill in the art to make transition metal oxides which sustain a superconducting current at T  $_{\rm c} > 26\,^{\rm o}$ K. The Examiner has provided no factual evidence that Applicants claims are not enabled for transition metals.

### The Examiner states:

iii. The applicants argue that their own examples do not support the determination of non-enabling scope of the invention. Nevertheless, the record is viewed as a whole. If the applicants could not show superconductivity with a Tc > 26°K for certain compositions falling within the scope of the present claims, it is unclear how someone else skilled in the art would have been enabled to do so at the time the invention was made.

The Examiner incorrectly states "applicants could not show superconductivity with  $T_c > 26\,^{\circ}\text{K}$  for certain compositions falling within the scope of the present claims." The present claims are directed to a method of flowing a superconducting current in a composition having a  $Tc > 26\,^{\circ}\text{K}$ . If a composition has a  $Tc < 26\,^{\circ}\text{K}$ , a method of flowing a superconducting current in such a compound cannot fall within the scope of applicants claims. Applicants are not claiming a composition of matter. They are claiming their discovery, passing a superconductive current through a transition metal oxide having a  $Tc > 26\,^{\circ}\text{K}$ . No one prior to applicants knew this. That is why they received the Nobel Prize in Physics in 1987.

The Examiner further states that Applicants' affidavits are conclusory. The Examiner appears to be placing himself up as an expert in the field of superconductivity. Applicants requested that the Examiner submit an affidavit in the present application rebutting the position taken by applicants' five affiants, but the Examiner has not submitted an affidavit. The facts are that the five affiants are experts in the art, the Examiner is not. The Examiner states that those "affidavits were insufficient because they were conclusory only, i.e., they lacked particular facts to support the conclusions reached". Applicants submitted the affidavit of Peter Duncombe which has provided hundreds of pages of notebook entries showing that he fabricated superconductive transition metal oxides according to the teaching of applicants specification.

The Examiner has provided no substantial evidence to support this assertion of non-enabling scope of the invention. It is requested that the Examiner support his assertion with factual evidence and not unsupported statements.

The Examiner is applying an incorrect standard of enablement. The Examiner is applying a standard applicable to composition of matter. Applicants are not claiming a composition of matter. As shown by applicants prior comments applicants have in fact fully enabled the composition of matter. Therefore, applicants have provided excess enablement for the claimed invention. The standard of enablement for a method of use is not the same as the standard of enablement for a composition of a matter. Notwithstanding, it is well settled law that claims to a composition of matter can encompass a number of inoperable species. However, Applicants' claims do not cover any inoperable species. The claims only encompass methods of flowing a superconducting current in transition metal oxides that are superconducting at temperatures in excess of 26°K. Those transition metal oxides that are not superconducting at temperatures in excess of 26°K are not encompassed by applicants claims reciting these limitations. Applicants note that a claim to a composition of matter is dominant to any use of that composition of matter and claims directed to a method of use of a composition of matter are necessarily of narrower scope than claims to the composition of matter. Applicants' claims do not encompass uses other than those which the claims are limited to by the use limitations recited in the claims. Applicants' claims are directed to what they have discovered. Therefore, applicants' claims fully satisfy the requirements of 35 USC 112.

The claimed invention is enabled because it is directed to a method of use rather than a composition. Applicants are claiming their discovery, showing superconducting current in a transition metal oxides with a Tc > 26°K. If a patent applicant claims a method of flowing current through a circuit having a resistive element, the applicant does not have to describe every method of making every type of resistive element for the claim to dominate all resistive elements. Such a claim reads on resistive elements made of materials not known at the time of filing since the discovery is not the material but the method of use. Applicants discovered that a superconducting current can be flowed in a transition metal oxides having a Tc > 26°K. That is what applicants are claiming.

Process of use claims are subject to the statutory provisions of 35 USC 112, first paragraph. All that is necessary to satisfy §112 is the statement that a superconducting current can be passed through a transition metal oxides having a Tc > 26 °K. The Examiner has essentially said this by rejecting applicants non-allowed claims as anticipated under §102(a) or obvious under §103(a) in view of the Asahi Shinbum article. Applicants only allowed claim 136 was allowed over the Asahi Shinbum article because it showed criticality for the formula recited in claim 136. Since Applicants generic teaching does not prevent others from obtaining patents to specific formulas, Applicants are entitled to generic claims to their discovery. Applicants filed this application soon after their discovery. Applicants availed themselves of the one year grace period under 35 USC 102(b) by publishing their results before filing the present

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application. This was the quickest way to promote the progress of the field of high T<sub>c</sub> superconductivity which can have substantial societal benefits such as less expensive electric power and more effective medical diagnostic tools. It is a policy of the United States Constitution, which establishes the United States Patent System, to encourage early disclosure of inventions to promote the progress of the useful arts. The Examiner's position that Applicants generic claims are not fully enabled frustrates this policy. Applicants could have decided not to publish Applicants' article and not to file the present application while engaging in years of further experimentation to find all specific examples which had the optimal T<sub>c</sub>. If Applicants acted this way, there would not have been the explosive worldwide effort to fully explore and implement high T<sub>c</sub> transition metal oxide technology. The rationale used by the Examiner is contrary to the Constitutional policy to promote the progress of the useful arts by early disclosure of an invention. Early disclosure should not be a penalty to Applicants. Applicants are pioneers in discovering that transition metal oxides have Tc > 26°K. A first discoverer of a wheel whose specific embodiment is a solid disc rotateable about an axle can claim a clylindrical member adapted for rotation about the axle and for rolling on a surface, that is, their discovery. This claim is dominant to a latter inventor's improved wheel comprising spokes which has the advantage of much lighter weight than a disc. The latter inventor is entitle to subservient claim to the dominant claim to a wheel. Applicants are entitled to a dominant claim to their discovery.

The Examiner has not shown by evidence not contained within applicants' teaching that the art of high Tc superconductors is unpredictable. The Examiner has

attempted to use applicants' teaching to establish such unpredictability. The Examiner has essentially not responded to Applicants' comments. The Examiner has introduced no extrinsic evidence to support the Examiner's opinion that the field of high  $T_c$  superconductivity is unpredictable. The Examiner has not introduced an affidavit qualifying himself to offer such an opinion. The Examiner merely states that his opinion is supported by the record as a whole. The only thing that Examiner relies on is some examples referred to above which do not have  $T_c > 26$ °K which as stated above is part of Applicants' enabling disclosure.

In response to the resubmitted 1.132 Affidavits of Mitzi, Tsuei and Dinger (filed 12/16/98) and the newly provided 1.132 Affidavits of Thomas Shaw (filed 12/16/98) and Peter Duncombe (filed 12/21/98) the Examiner states:

The Examiner notes the books describing the general principles of ceramic fabrication provided in these Affidavits. Also, the examiner notes that such fabrication techniques were utilized subsequently (after the filing date of the instant application) to produce the superconductive materials described in the Poole et al reference as well as the materials produced by Peter Duncombe (see Affidavit filed 12/21/98).

The affidavit of Peter Duncombe provides notebook entries beginning November 12, 1987, which is prior to Applicants filing date and prior to the Asahi Shinbum article.

Duncombe used the general principles of ceramic science to implement the invention

described in Applicants article. Each of the five affiants Tsuei, Mitzi, Duncombe, Shaw and Dinger are experts in the filed of ceramics. Each has stated the Applicants' claimed invention can be implemented based on Applicants teaching and with nothing more than the general principles of ceramic science known at the time of filing of the present application.

## The Examiner states:

It is the examiner's maintained position that while general principles of ceramic fabrication were most certainly known prior to the filing date of the instant application, the utilization of such techniques to produce superconductive materials within the scope of the instant claims were not known. The affidavits are not effective to demonstrate enablement at the time of the invention was made. As stated in paper #66, page 8, one may now know of a material that superconduct at more than 26K, but the affidavits do not establish the existence of that knowledge on the filing date of the present application.

The Examiner acknowledges that the fabrication techniques necessary to practice Applicants' invention were known prior to the filing dated of the present application. But the Examiner further states that the "utilization of such techniques to produce superconductive materials within the scope of the instant claims were not known". The scope of the instant claim is flowing a superconductive current in a

transition metal oxide having a  $T_c > 26$ °K. That is Applicants' discovery. That is why it was not known prior to Applicants' discovery. How to make this type of material was known. Prior to Applicants' discovery, It was not known that they were superconductive with a  $T_c > 26$ °K. The Examiner incorrectly states "one may now learn of a material that superconducts at more than 26°K, but the affidavits do not establish the existence of that knowledge on the filing date of the present invention." If that knowledge was known by another prior to the filing date, Applicants would not have a patentable invention since they would not be the initial first and sole inventor. The affidavits state that the knowledge of how to make transition metal oxides by the general principles of ceramic science were known prior to the filing date. In particular, the affidavits of Duncombe and Shaw refer to a number of articles and texts on the general principles of ceramic science. One of these texts is "Structures, Properties and Preparation of Peroskite-type Compounds", F.S. Galasso (1969).

Applicants note that the book "Copper Oxide Superconductors" by Charles P. Poole, Jr., Timir Datta and Horacio A. Farach, John Wiley & Sons (1998) support their position that high temperature superconductors were not difficult to make after their original discovery. This book shall be referred to herein as Poole et al. or the Poole book. The Poole book was published after Applicants' initial discovery which was published in Applicants' article. The Examiner states "[a]s such, it does not, provide evidence of the state of the art at the time the presently claimed invention was made".

Applicants disagree. The preface of this book says "[t]his volume reviews the experimental aspects of the field of oxide superconductivity with transition temperatures from 30K to above 123K, from the time of its discovery by Bednorz and Muller in April. 1986 until a few months after the award of the Nobel Prize to them in October, 1987." Thus the book reports on work done within eighteen months of Applicants' discovery in April 1986 and within eleven months of its publication in September, 1986. In the present application was filed on May 22, 1987. This passage is referring to Applicants and Applicants' article referred to at page 6 of Applicants' specification. This book acknowledges that applicants are the discovers of the field of high temperature superconductivity. (See Attachment A of Applicants' response dated May 14, 1998 and See Attachment H of Applicants' response dated November 28, 1997). The Examiner's view that the skill of the art was insufficient at the time of the filing date of the present application is untenable in the view of Poole et al. and Applicants' 132 affi davits of Tsuei, Mitzi, Shaw, Dinger and Duncombe, in particular that of Peter Duncombe which reports data prior to the Applicants' filing date.

Applicants note that it is generally recognized that it is not difficult to fabricate transition metal oxides and in particular copper metal oxides that are superconductive after the discovery by applicants that transition metal oxides are high T<sub>c</sub> superconductors. Chapter 5 of the Poole et al. book entitled "Preparation and Characterization of Samples" states at page 59 "[c]opper oxide superconductors with a purity sufficient to exhibit zero resistivity or to demonstrate levitation (Early) are not difficult to synthesize. We believe that this is at least partially responsible for the

explosive worldwide growth in these materials". Poole et al. further states at page 61 "[i]n this section three methods of preparation will be described, namely, the solid state, the coprecipitation, and the sol-gel techniques (Hatfi). The widely used solid-state technique permits off-the-shelf chemicals to be directly calcined into superconductors, and it requires little familiarity with the subtle physicochemical process involved in the transformation of a mixture of compounds into a superconductor." Poole et al. further states at pages 61-62 "[i]n the solid state reaction technique one starts with oxygen-rich compounds of the desired components such as oxides, nitrates or carbonates of Ba, Bi, La, Sr, Ti, Y or other elements. ... These compounds are mixed in the desired atomic ratios and ground to a fine powder to facilitate the calcination process. Then these room-temperature-stabile salts are reacted by calcination for an extended period (~20hr) at elevated temperatures (~900°C). This process may be repeated several times, with pulverizing and mixing of the partially calcined material at each step." This is generally the same as the specific examples provided by applicants and as generally described at pages 8, line 19, to page 9, line 5, of Applicants' specification which states "[t]he methods by which these superconductive compositions can be made can use known principals of ceramic fabrication, including the mixing of powders containing the rare earth or rare earth-like, alkaline earth, and transition metal elements, coprecipitation of these materials, and heating steps in oxygen or air. A particularly suitable superconducting material in accordance with this invention is one containing copper as the transition metal." (See Attachment A of Applicants' response dated May 14, 1998 and See Attachment H of Applicants' response dated November 28, 1997).

Consequently, applicants have fully enabled high Tc transition metal oxides and their claims.

As stated in the affidavit of Dr. Mitzi, Dr. Dinger, Dr. Tsuei, Dr. Shaw and Mr. Doncombe the preface of the book by Poole et al., quoted above, the work of Applicants initiated the field of high temperature superconductors and these materials are not difficult to synthesize. And according In re Fisher "it is apparent that such an inventor should be allowed to dominate future patentable inventions of others where those inventions were based in some way on his teaching." (166 USPQ 18, 24)

#### The Examiner further states:

Moreover, the present claims are directed to processes of using metal oxide superconductors, not processes of making them. Even if the Poole article provided direct evidence of the state of the art at the time the invention was made, which it apparently does not, that evidence still does not pertain to the issue at hand, namely, the process of using metal oxide superconductors to conduct electricity under superconducting conditions.

Applicants disagree. Poole et al. Chapter **X** entitled "Transport Properties" describes a process of using metal oxides superconductors to conduct electricity under superconducting conditions. Section B of Chapter **X** is entitled "Current Flow". A copy of Chapter **X** is in Attachment B of this Appeal Brief.

The Examiner further states in regard to Poole et al.:

(3) Finally, the Preface states in part at A3: "The unprecedented worldwide effort in superconductivity research that has taken place over the past two years has produced an enormous amount of experimental data on the properties of the copper oxide type materials that exhibit superconductivity above the temperature of liquid nitrogen. ... During this period a consistent experimental description of many of the properties of the principal superconducting compounds such as BiSrCaCuO, LaSrCuO, TIBaCaCuO, and YBaCuO has emerged. ... The field of high-temperature superconductivity is still evolving ..." That preface is deemed to show that the field of high-temperature superconductivity continued to grow, on the basis of on-going basic research, after the Bednorz and Meuller article was published.

The continued growth referred to in the passage from Poole et al. quoted above does not mean that this work is not based on Applicants' initial fundamental teaching. Poole et al. as quoted above states that the unprecedented amount of work done in the short period of time after Applicants' work was because the materials "are not difficult to synthesize." Moreover, as quoted above the CCPA In re Fisher states "such an inventor should be allowed to dominate future patentable inventions of other where those inventors were based on in some way on his teachings." Moreover, the referred

to future developments in the passage above are not necessarily patentably distinct from Applicants' teachings. Those who developed these compounds would have a reasonable expectation of success based on Applicants' teaching. The Examiner has provided no evidence to the contrary.

The Examiner acknowledges that the three affidavits of Dr. Tsuei, Dr. Dinger and Dr. Mitzi which were signed in May of 1998 states that the present application "includes all known principles of ceramic fabrication known at the time the application was filed." However, the Examiner further states that the "additional indication also is considered to be a conclusory statement unsupported by particular evidence". The advisory action dated February 25, 1998 acknowledges the revised 1.132 affidavits of Mitzi, Tsuei and Dinger (filed 12/16/98) and the newly provided affidavit of Shaw (filed 12/16/98) and Dumcombe (filed 12/21/98). These revised and additional affidavits cite numerous articles and books, all published before the Applicants' filing date, which provide evidence of the general teaching of ceramic science, and in particular, ceramic fabrication techniques for transition metal oxides, more particularly for Perovskites and most particularly for the type of Perovskites of Applicants' preferred embodiments. Applicants disagree that their affidavits are conclusory. The facts are provided by Poole et al. The affidavits corroborate what Poole et al. states.

Claims 134, 135, and 137-142 were rejected under 35 U.S.C. 112, first paragraph, as based on a disclosure which is not enabling. The Examiner states "[e]ach of claims 134, 135, and 137-142 provide for a superconductor "having a Tc

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greater than 26°K", but those claims do not provide for a step of -- maintaining said (superconductor) at a temperature less than said Tc". These claims have been amended by the amendment after final rejection dated June 14, 1999, to include the steps of maintaining the appropriate temperature for superconductivity as suggested by the Examiner.

In view of these arguments the Board is requested to reverse the Examiner's rejection of claims 24-26, 86-90, 96-113, 129-131, 134, 135-166, 169, 170 and 174-177 as not enabled under 35 USC 112, first paragraph.

# **REJECTIONS UNDER 35 USC 112, SECOND PARAGRAPH**

Claims 86-87, 96-108, 115, 118, 120, 122, 123, 129-135, and 137-142 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

In paragraph 6-7 on page 12-16 of the final rejection, Claims 86-87, 96-108, 115, 118, 120, 122, 123, 129-135 and 137-142 have been rejected under 35 USC 112. All changes suggested by the examiner have been made by Amendment after Final Rejection dated June 14, 1999, except for those directed to the terms "layer-like", "perovskite-like", "rare-earth-like", and "layer-type". These terms occur in claims 86-87, 96-108, 112, 113, 117, 118, 122 and 123. As stated by the applicants in the prosecution these are terms of art and well understood by persons of skill in the art.

Claims 143-163 correspond to in claims 86-87, 96-108, 112, 113, 117, 118, 122 and 123. Claims 143-163 have the same wording as claims 86-87, 96-108, 112, 113, 117, 118, 122 and 123 and include all changes suggested by the examiner to overcome rejections of claims 86-87, 96-108, 112, 113, 117, 118, 122 and 123 claims under 35 USC 112, second paragraph.

The Examiner states:

- a. With respect to claims 86-87 and 96-108, the terms "layer-type","perovskite-like", "rare-earth-like" are vague and confusing.
- i. The question arises: What is meant by these terms?
- (1) The terms "layer-type" and "perovskite-like" are unclear because the "type" or "like" terms are deemed to be indefinite. Terms such as "like", "similar", and "type" are indefinite." It is suggested that "layer-type perovskite-like crystal structure" be changed -- a substantially layered perovskite crystal structure --.

In support of the Examiner's view of term such as "type" and "like", the Examiner cites: Ex parte Remark, 15 USPQ 2d 1498, 1500 (BPAI 1990); Ex parte Kristensen, 10 USPQ 2d 1701, 1703 (BPAI 1989); Ex parte Attig, 7 USPQ 2d 1092, 1093 (BPAI 1988); and Ex 'parte Copenhaver, 109 USPQ 118 (POBA 1955).

Ex parte Remark is not on point since the Board found the term "and the like" in a claim to be indefinite under §112. Applicants claims do not use this terminology.

Ex parte Kristensen is not on point since the Board found the term "similar" in the phase "for high pressure cleaning units or similar apparatus" in a claim indefinite under §112, second paragraph. Applicants do not use the terminology "or similar". The Board held with respect to the second paragraph of §112, the inquiry is "to determine

whether the claims do, in fact, set out and circumscribe a particular area with a reasonable degree of precision and particularity". In the present application the terms "perovskite-like", "perovskite-type", "layered-like", "layered-type" and "rare-earth-like" are terms used in the art and these are well known and understood by a person of skill in the art.

Ex parte Attig is also not on point. The Board found the term "ZSM-5-type" indefinite under §112, second paragraph, because the prior art cited during prosecution gave different meanings to the term "ZSM-5-type". But the Board did not find the use of the term "type" in a claim per se indefinite. The Board says "[i]t is true that the zeolites have been defined in various patents and claimed with the terminology "ZSM-5-type". However, clearly the art of record in this case, all of it cited by applicants themselves, in the aggregate serve to render the term indefinite rather that definite". 7 USPQ 1092, 1093. The Examiner has not shown that the terms "perovskite-like", "perovskite-type", "layered-like", "layered-type" and "rare-earth-like" as used in applicants claims have different meanings in different cited prior art. To the contrary, Applicants have shown that these terms have a uniform meaning in the transition metal oxide high Tc art.

In Ex parte Copenhaver, the Board stated in regard to the terminology "Friedel-Graftz type" catalyst.

"[w]e are of the view that the word "type" when appended to another wise definite expression so extends the scope of such an expression as to render it objectionably indefinite from the stand point of patent law and procedures."

We are not led to a different conclusion by the fact that the expression may have been used in certain technical and scientific publications which are not subject to the rigid legal requirements for definitions that apply to patent claims. The fact that the expression may have been used in claims of certain patents likewise does not alter our view on the question."

Applicants note that Ex parte Copenhaver decided in 1955 was not cited by the Board in the later Board decisions cited by the Examiner. This decision has not been cited by any other Board or Court decision.

The Examiner states that the term "rare earth-like element" is indefinite and that the terms "type and "like" are unclear. Applicants respectfully disagree. Attachment A of Applicants' response dated November 28, 1997 is the result of a Lexis search done by the undersigned attorney. This search shows the term "rare earth like" or "rare earth and the like" used in 68 US patents. The sections of the 68 patents where these terms appears are printed using the "KWICK" function of Lexis. Attachment B of Applicants' response dated November 28, 1997 is the results of a Lexis search done by the undersigned attorney. This search shows the term "rare earth like" used in the claims of 4 issued US patents. The sections of the claims of the 4 patents where this term appears in the claims are printed using the "KWICK" function of Lexis. Consequently,

the term "rare earth like" is a term use in the art, understood by a person of skill in the art and recognized as a definite term by the USPTO for use in US patent claims.

The Examiner states that the term "perovskite-like" is indefinite. The term "perovskite-like" or "perovskite-type" is commonly used in the art. Attachment C of Applicants' response dated November 28, 1997 is the results of a Lexis search done by the undersigned attorney. This search shows that the terms "perovskite like" and "supercond!" (the "!" represents any combination of letters) are used in 107 US patents. The section of the 107 patents where these terms appears are printed using the "KWICK" function of Lexis. Attachment D of Applicants' response dated November 28,1997 is a result of a Lexis search done by the undersigned attorney. This search shows the terms "perovskite like" or 'perovskite type" used in the claims of two issued US patents. The sections of the claims of the 2 patents where this term appears in the claims are printed using the "KWICK" function of Lexis. Attachment E of Applicants' response dated November 28, 1997 is a copy of the first page of Chapter 2 of the book "Perovskites and High Tc Superconductors" by F. S. Galasso, Gordon and Breach Scientific Publishers, 1990. Chapter 2 is entitled "Structure of Perovskite-type Compounds". Attachment F of Applicants' response dated November 28, 1997 is a copy of page 78 of the book by C. Poole, Jr. et al. Page 78 is the beginning of the section entitled "D. Perovskite-type Superconducting Structures". The first paragraph of the section states "[i]n their first report on high-temperature superconductors Bednorz and Muller (the applicants) referred to their samples as 'metallic, oxygen deficient ... perovskite like mixed valent copper compounds.' Subsequent work has confirmed that

the new superconductors do indeed have these characteristics. In this section we will comment on their perovskite-like aspects" (insert added). Consequently, the terms "perovskite like" or "perovskite type" are terms used in the art and recognized as a definite by the USPTO for use in US patent claims. (It is noted that this passage also shows that the terminology "mixed valent copper compounds" is used and understood in the art. Applicant further notes that one of the books cited in the affidavit of Peter Duncombe to demonstrate the general teaching of ceramic science includes in the title "Perovskite-Type Compounds". This book was published in 1969 showing that this term was well known and understood by those of the skill in the art long before the filing date of the present application.

The Poole et al. in Chapter VI on "Crystallographic Structures" states at page 73 "[m]uch has been said about the oxide superconductor compounds being **perovskite types**, so we will begin with a description of the perovskite structure." (emphasis added) (See Attachment I of Applicants response dated November 28, 1997).

The undersigned attorney did a search in Lexis for the terms "like" in issued US patents. As shown in Attachment A of Applicants' response dated December 27, 1997, this search Lexis found 140,058 issued US patents using the terminology "like" in the claims. The USPTO has, therefore, accepted this terminology as definite within the meaning of 35 USC 112. A number of specific examples are provided in Attachments B of Applicants' response dated December 27, 1997, to Attachment J of Applicants'

response dated December 27, 1997, each of which are the results of a similar Lexis search. (Lists of the patent numbers will be provided at the request of the Board.) Attachment B shows that there are 443 issued US patents having the term "diamond like" in the claims. Attachment C of Applicants' response dated December 27, 1997, shows that there are 319 issued US patents having the term "diamond like carbon" in the claims. Attachment D of Applicants' response dated December 27, 1997, shows that there are 10 issued US patents having the term "halogen like" in the claims. Attachment E of Applicants' response dated December 27, 1997, shows that there are 11 issued US patents having the term "oxygen like" in the claims. Attachment F of Applicants' response dated December 27, 1997, shows that there are 79 issued US patents having the term "ceramic like" in the claims. Attachment G of Applicants' response dated December 27, 1997, shows that there are 31 issued US patents having the term "carbon like" in the claims. Attachment H of Applicants' response dated December 27, 1997, shows that there are 5 issued US patents having the term "silicon like" in the claims. Attachment I of Applicants' response dated December 27, 1997, shows that there are 10 issued US patents having the term "nitrogen like" in the claims. Attachment J of Applicants' response dated December 27, 1997, shows that there are 17 issued US patents having the term "copper like" in the claims. In view thereof applicants respectfully request the Board to withdraw the rejection of their claims as indefinite for using the terminology "layer-like", "perovskite like" and "rare-earth-like" since use of the term "like" is recognized as definite under 35 USC 112 by the USPTO.

Also the term "ceramic-like" is a term commonly used in the art. Attachment M of Applicants' response dated November 28, 1997 is the result of a Lexis search performed by the undersigned attorney using the search criteria "ceramic" with one word of "like" and "copper" within one word of "oxide" and "rare" within one word of "earth". This search identified 23 issued US patents. These patents are listed in the attachment using the Lexis KWICK feature which list only those portions of the patents where these terms appear. The search was limited to this criteria since a search on "ceramic" within one word of "like" identified more than 1,000 issued US patents and a search on "ceramic" within one word of "like: in the same document as "copper" within one word of "oxide" identified more than 1000 US patents. It is clear that the term "ceramic like" is well understood in the art and is thus definite.

An affidavit under 37 CFR 1.132 of Dr. James W. Leonard, IBM research librarian was submitted on December 15, 1998. Dr. Leonard did a search of articles referencing Applicants article J. G. Bednorz and K. A. Muller, Zeitschrift fur Physik B-Condensed Matter, **64**, pp. 189-193 (Sept. 1986) which is incorporated by reference in the specification at page 6, lines 8-10. As stated at page 6 this article is the basis for applicants invention. More than 5800 articles have referenced applicants' article. This is evidence that applicants' teaching in the present application has motivated persons of skill in the art to work in the field of high T<sub>c</sub> superconductivity and that applicants teaching has fully enabled the invention of their claims. All these articles citing applicants' article acknowledge that their work is based on Applicants' teaching.

Claims 112, 113, 117, 118, 122 and 123 have been rejected under 35 USC 112 as indefinite for using terminology "layer-type". Applicants respectfully disagree.

The undersigned attorney did a search in Lexis for the terms "layer-type" and the term "layer" preceding the term "type" by one word (layer pre/1 type). In this search Lexis found 225 issued US patents using this terminology in the claims. The USPTO has, therefore, accepted this terminology as definite within the meaning of 35 USC 112. Attachment A of the Applicants' response dated December 22, 1998, is the results of this search printed out using the .kw or "kwick" feature which prints the searched term and words before and after the searched term. The searched terminology appears in a number of forms such as "layer type", "layer-type", "layer of type". In view thereof applicants request the Board to reverse the rejection of claims 112, 113, 117, 118, 122 and 123 under 35 USC 112 as indefinite for using the terminology "layer-type".

In attachment B of the Applicants' response dated December 22, 1998, there is a search done by Jim Leonard, an IBM Research librarian, at the request of the undersigned attorney of the term "Layered like" and "layered type" in on-line non-patent literature prior to applicants' filing date. From Attachment B of the Applicants' response dated December 22, 1998, it is clear that these terms are used and understood by persons of skill in the art.

In attachment C of the Applicants' response dated December 22, 1998, there is a search done by Jim Leonard, an IBM Research librarian, at the request of the

undersigned attorney of the term "rare earth like" in on-line non-patent literature prior to applicants' filing date. From this Attachment C it si clear that this term is used and understood by persons of skill in the art. This is in addition to the other evidence of the definiteness of this term to overcome the rejection of applicants claims as indefinite for using the term "rare earth like" which applicants respectfully request the Board to reverse

In Attachment D of the Applicants' response dated December 22, 1998, there is a search done by Jim Leonard, an IBM Research librarian, at the request of the undersigned attorney of the term "perovskite like" in on-line non-patent literature prior to Applicants' filing date. From this Attachment D it is clear that this term is used and understood by persons of skill in the art. This is in addition to the other evidence of the definiteness of this term to overcome the rejection of Applicants' claims as indefinite for using the term "perovskite like" which applicants respectfully request the Board to reverse.

The Examiner Further states:

(1) The terms "layer-type" and "perovskite-like" are unclear because the "type" or "like" terms are deemed to be indefinite. Terms such as "like", "similar", and "type" are indefinite." It is suggested that "layer-type perovskite-like crystal structure" be changed -- a substantially layered perovskite crystal structure --.

Applicants have said that person of skill in the art would understand (rare earth-like) to mean that a location occupied by a rare earth element can also be occupied by another element which would have chemical properties similar enough to the rare earth elements such that it would fit in to the lattice site occupied by the rare earth element and act chemically as a rare earth element. To this the Examiner has responded "[t]hat response does not alleviate the problem, however. Other elements may 'fit' into the lattice but they may not necessarily be 'rare-earth-like'". This comment does not address the issue. These are only about 100 elements of which 14 or 15 are rare-earth elements leaving about 86 other elements. It is not difficult for a person of skill in the art to determine which elements will behave like a rare-earth element when placed in the lattice.

Claims 112, 113, 115, 117, 118, 120, 122, and 123 have been found to be indefinite for the reasons that follow.

- i. In claim 112, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.
- ii. In claim 113, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.
- iv. In claim 117, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.

- v. In claim II 8, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.
- viii. In claim 122, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.
- ix. In claim 123, line 5, has the terms "layer-type" and "perovskite-like", both of which are indefinite.
- d. Applicants' arguments filed March 7, 1997 (#59) have been fully considered but they are not persuasive.

These issues have been addressed above.

Applicants have cited numerous issued U.S. patents using "type" and "like" in the claims. Many of these patents used the identical terminology objected to by the Examiner which has been used by Applicants. Many issued U.S. patents similar terminology to that used by Applicants. The Examiner has provided no reason for rejecting Applicants claims using the terminology "layer-type", "perovskite like", "perovskite type" and "rare earth like" under §112, second paragraph as indefinite when there are issued patents using these and related terms in the claims.

In view of these arguments applicants request the Board to reverse the rejection of claims 86-87, 96-108, 115, 118, 120, 122, 123, 129-135, 137-142, 164-169 and 170-177 as being indefinite under 35 U.S.C. § 112, second paragraph.

## SPECIFIC ARGUMENTS AS TO EACH CLAIM

In this section of this appeal brief the Asahi Shinbum article is referred to as ASA. The expression "the Asahi Shinbum article provides no teaching suggestion, motivation for or incentive for the limitations highlighted in the claim language above" is abbreviated as "ASA provides no TSMA for the limitation highlighted in the claim language above". In the rejection of all of applicants claims, except for claim 136, for no enablement under §112, first paragraph, the Examiner has not specifically referred to any of the rejected claims in paragraphs c to f on pages 6 to 12 which is the part of the final rejection directed to the claim rejects under §112, first paragraph. In the objection to the specification under §112, first paragraph the Examiner only refers to claims 24, 88, 96 and 103. Applicants' comments as to dependent claims include the Applicants' comments as to the claims from which they depend even if not repeated.

The best mode contemplated at the time the present application was filed was the subject of claim 136. Support for each of the claims and how a person of skill in the art can make and use each claim is described above.

#### Claim 24 recites:

24. A method including the steps of forming a transition metal oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K,

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase, and

passing an electrical supercurrent through said transition metal oxide while it is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 25 recites:

25. The method of claim 24, where said transition metal oxide is comprised of a transition metal capable of exhibiting **multivalent states**.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

### Claim 26 recites:

26. The method of claim 24, where said transition metal oxide is comprised of a Cu oxide.

See comments for claim 24.

#### Claim 86 recites:

86. A method, comprising the steps of:

forming a composition including a transition metal, a rare earth or rare earth-like element, an alkaline earth element, and oxygen, where said composition is a mixed transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K,

maintaining said composition in said superconducting state at a temperature greater than 26°K, and

passing an electrical current through said composition while said composition is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot

practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art. As to "like" or "type" terminology being indefinite under §112, second paragraph see the discussion above.

#### Claim 87 recites:

87. The method of claim 86, where said transition metal is copper. See the discussion for claim 86.

## Claim 88 recites

88. A method, including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K, maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state, and

passing an electrical current through said composition while said composition is in said superconductive state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot

practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

### Claim 89 recites:

89. The method of claim 88, where said composition is comprised of a metal oxide.

See the comments for claim 88.

# Claim 90 recites:

90. The metal of claim 88, where said composition is comprised of a transition metal oxide.

See the comments for claim 88.

#### Claim 96 recites:

- 96. A superconductive method for causing electric current flow in a superconductive state at a temperature in excess of 26 K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductor transition

temperature T<sub>c</sub> of greater than 26 K;

- (b) maintaining the superconductor element at a temperature above 26 K and below the superconductor transition temperature  $T_{\rm c}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

#### Claim 97 recites:

97. The superconductive method according to claim 96 in which the copper-oxide compound of the superconductive composition includes at least one rare-earth or rare-earth-like element and at least one alkaline-earth element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot

practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

### Claim 98 recites:

98. The superconductive method according to claim 97 in which the rare-earth or rare-earth-like element is lanthanum.

See comments for claim 97.

### Claim 99 recites:

99. The superconductive method according to claim 97 in which the alkaline-earth element is barium.

See comments for claim 97.

## Claim 100 recites:

100. The superconductive method according to claim 96 in which the copper-oxide compound of the superconductive composition includes **mixed valent copper ions**.

In addition to the comments on claim 96, as the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 101 recites:

101. The superconductive method according to claim 100 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.

In addition to the comments on claim 100. As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 102 recites:

102. The superconductive method according to claim 101 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.

In addition to the comments on claim 101, as the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

# Claim 103 recites:

- 103. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a

copper-oxide compound having a **layer-type** perovskite-like crystal structure, the copper-oxide compound **including at least one rare-earth** or rare-earth-like element and at least one alkaline-earth element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than 26 K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

### Claim 104 recites:

104. The superconductive method according to claim 103 in which the rare-earth or rare-earth-like element is lanthanum.

See comments on claim 103.

# Claim 105 recites:

105. The superconductive method according to claim 103 in which the alkaline-earth element is barium.

See comments on claim 103.

#### Claim 106 recites:

106. The superconductive method according to claim 103 in which the copper-oxide compound of the superconductive composition includes **mixed valent copper ions**.

#### Claim 107 recites:

107. The superconductive method according to claim 106 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

#### Claim 108 recites:

108. The superconductive method according to claim 107 in which oxygen is present in the copper-oxide compound in a **nonstoichiometric** atomic proportion.

## Claim 109 recites:

109. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

### Claim 110 recites:

110. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

### Claim 111 recites:

111. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and an element selected from the group consisting of Group II A element, a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 112 recites:

112. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, a rare earth element; and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}\text{K}$  and below the superconductor transition temperature  $T_{\circ}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

### Claim 113 recites:

- 113. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T<sub>p=0</sub>, the transition-onset temperature T<sub>c</sub> being greater than 26°K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

  As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph

the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

### Claim 114 recites:

114. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

#### Claim 115 recites:

115. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 116 recites:

116. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26 °K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A and at least one element selected from the group consisting of a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

## Claim 117 recites:

117. A superconductive method for causing electric-current flow in a superconductive state at a temperature **in excess of 26°K**, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}\text{K}$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

# Claim 118 recites:

118. A superconductive method for conducting an electric current essentially without resistive losses, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a **layer-type** perovskite-like crystal structure, the copper-oxide compound **including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element,** the composition having a superconductive/resistive transition defining a superconductive-resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^{\circ}K$ ;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

  As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

### Claim 119 recites:

119. A method including the steps of forming a transition metal oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said transitional metal oxide includes at least one element selected from the group consisting of a Group II A element and at lest one element selected from the group consisting of a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

## Claim 120 recites:

120. A method comprising the steps of:

forming a composition including a transition metal, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed transitional metal oxide formed from said transition metal and said oxygen, said mixed transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

#### Claim 121 recites:

121. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26 °K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a transitional metal oxide and at least one element selected from the group consisting of Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

# Claim 122 recites:

- 122. A superconductive method for causing electric-current flow in a superconductive state at a temperature **in excess of 26°K**, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a

transition metal oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;

- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}\text{K}$  and below the superconductor transition  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

  As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

### Claim 123 recites:

- 123. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal-oxide compound having a layer-type perovskite-like

one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T p=0, the transition-onset temperature T<sub>c</sub> being greater than 26°K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

### Claim 124 recites:

124. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature **in excess of 26°K**;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 125 recites:

125. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element at least one element selected from the group consisting of a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 126 recites:

126. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26 °K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 127 recites:

- 127. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting

essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element;

- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}K$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

### Claim 128 recites:

- 128. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a

copper-oxide compound having a **layer-type** perovskite-like crystal structure, the copper-oxide compound including **at least one element selected from the group consisting of a group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element, the composition having a superconductive-resistive transition temperature defining a** superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^o$ K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above.

## Claim 129 recites:

129. A method comprising providing a composition having a transition temperature **greater than** 26°K, the composition including a rare earth or alkaline earth element, a transition metal element capable of exhibiting **multivalent states** and oxygen, including at least one phase that exhibits superconductivity at temperature in excess of 26°K, maintaining said composition at said temperature to exhibit said superconductivity and passing an electrical superconducting current through said composition with said phrase exhibiting said superconductivity.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 130 recites:

130. A method comprising providing a superconducting transition metal oxide having a superconductive onset temperature **greater than 26°K**, maintaining said superconducting transition metal oxide at a temperature less than said superconducting onset temperature and flowing a superconducting current therein.

## Claim 131 recites:

131. A method comprising providing a superconducting copper oxide having a superconductive onset temperature **greater than 26°K**, maintaining said superconducting copper oxide at a temperature less than said superconducting onset temperature and flowing a superconducting current in said superconducting oxide.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 132 recites:

132. A method comprising providing a superconducting oxide composition having a superconductive onset temperature **greater than 26°K**, maintaining said superconducting copper oxide at a temperature

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less than said superconducting onset temperature and flowing a superconducting current therein, said composition comprising at least one each of rare earth, an alkaline earth, and copper.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 133 recites:

133. A method comprising providing a superconducting oxide composition having a superconductive onset **temperature greater than**26°K, maintaining said superconducting copper oxide at a temperature less than said superconducting onset temperature and flowing a superconducting electrical current therein, said composition comprising at least one each of a Group III B element, an alkaline earth, and copper.

#### Claim 134 recites:

134. A method comprising flowing a superconducting electrical current in a transition metal oxide having a  $T_c$  greater than 26°K and maintaining said transition metal oxide at a temperature less than said  $T_c$ .

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 135 recites:

135. A method comprising flowing a superconducting current in a copper oxide having a  $T_c$  greater than 26°K and maintaining said copper oxide at a temperature less than said  $T_c$ .

# Claim 136 is allowed.

# Claim 137 recites:

137. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K, said composition comprising at least one each of a III B element, an alkaline earth, and copper oxide and maintaining said composition of matter at a temperature less than  $T_c$ .

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 138 recites:

138. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K, said composition comprising at least one each of a rare earth, alkaline earth, and copper oxide and maintaining said composition of matter at a temperature less than said  $T_c$ .

# Claim 139 recites:

139. A method comprising flowing a superconducting electrical current in a composition of matter having a T<sub>c</sub> greater than 26°K, said composition comprising at least one each of a rare earth, and copper oxide and maintaining said composition of matter at a temperature less than said T<sub>c</sub>.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 140 recites:

140. A method comprising flowing a superconducting electrical current in a composition of matter having a T<sub>c</sub> greater than 26°K carrying, said composition comprising at least one each of a III B element, and copper

oxide and maintaining said composition of matter at a temperature less than said  $\mathsf{T}_c$ .

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 141 recites:

141. A method comprising flowing a superconducting electrical current in a transition metal oxide comprising a  $T_c>26^\circ K$  and maintaining said transition metal oxide at a temperature less than said  $T_c$ .

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 142 recites:

142. A method comprising flowing a superconducting electrical current in a copper oxide composition of matter comprising a T<sub>c</sub>>26°K and

maintaining said copper oxide composition of matter at a temperature less than said  $\mathsf{T}_c$ .

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 143 recites:

143. A method, comprising the steps of:

forming a composition including a transition metal, a group IIIB element, an alkaline earth element, and oxygen, where said composition is a mixed transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K,

maintaining said composition in said superconducting state at a temperature greater than 26°K, and

passing an electrical current through said composition while said composition is in said superconducting state.

# Claim 144 recites:

144. The method of claim 143, where said transition metal is copper. See comments for claim 143.

# Claim 154 recites:

- 145. A superconductive method for causing electric current flow in a superconductive state at a temperature **in excess of 26 K**, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a **substantially layered** perovskite crystal structure, the composition having a superconductor transition temperature  $T_c$  of greater than 26 K;
- b) maintaining the superconductor element at a temperature above 26 K and below the superconductor transition temperature  $T_{\rm c}$  of the superconductive composition; and

# Claim 146 recites:

146. The superconductive method according to claim 145 in which the copper-oxide compound of the superconductive composition includes at least one element selected from the group consisting of a rare-earth element and a Group III B element and at least one alkaline-earth element.

# Claim 147 recites:

147. The superconductive method according to claim 146 in which the rare-earth or rare-earth-like element is lanthanum.

See the comments as to claim 146 and as to "like" or "type" terminology being indefinite under §112, second paragraph, see the discussion above.

## Claim 148 recites:

148. The superconductive method according to claim 146 in which the alkaline-earth element is barium.

See comments as to claim 46.

## Claim 149 recites:

149. The superconductive method according to claim 145 in which the copper-oxide compound of the superconductive composition includes **mixed valent copper ions**.

# Claim 150 recites:

150. The superconductive method according to claim 149 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 151 recites:

151. The superconductive method according to claim 150 in which oxygen is present in the copper-oxide compound in a **nonstoichiometric** atomic proportion.

# Claims 152 recites:

- 152. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a rare-earth element and a Group III B element and at least one alkaline-earth element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T<sub>p=0</sub>, the transition-onset temperature T<sub>c</sub> being greater than 26 K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

# Claim 153 recites:

153. The superconductive method according to claim 152 in which said at least one element is lanthanum.

See comments as to claim 152.

## Claim 154 recites:

154. The superconductive method according to claim 152 in which the alkaline-earth element is barium.

See comments as to claim 152.

# Claim 155 recites:

155. The superconductive method according to claim 152 in which the copper-oxide compound of the superconductive composition includes mixed valent copper ions.

# Claim 156 recites:

156. The superconductive method according to claim 155 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.

In addition to comments on claim 155, as the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 157 recites:

157. The superconductive method according to claim 156 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.

# Claim 158 recites:

- 158. A superconductive method for causing electric-current flow in a superconductive state at a temperature **in excess of 26°K**, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, a rare earth element; and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26\,^\circ K$  and below the superconductor transition temperature  $T_\circ$  of the superconductive composition; and

(c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 159 recites:

- 159. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a **substantially layered** perovskite crystal structure, the copper-oxide compound including **at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element**, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T<sub>p=0</sub>, the transition-onset temperature T<sub>c</sub> being greater than 26°K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

# Claims 160 recites:

- 160. A superconductive method for causing electric-current flow in a superconductive state at a temperature **in excess of 26°K**, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a

Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;

- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}K$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 161 recites:

- 161. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the copper-oxide compound including at least one element

selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive-resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^{\circ}K$ ;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 162 recites:

- 162. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature **above 26°K** and below the superconductor transition  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

## Claim 163 recites:

- 163. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal-oxide compound having a **substantially layered** perovskite crystal structure, the transition metal-oxide compound including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T p=0, the transition-onset temperature T<sub>c</sub> being greater than 26°K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

## Claim 164 recites:

164. A method according to claim 129 wherein said composition comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 165 recites:

165. A method according to claim 130 wherein said superconducting transistor metal oxide comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation

highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 166 recites:

166. A method according to claim 131 wherein said superconducting copper oxide comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 167 recites:

167. A method according to claim 132 wherein said superconducting oxide composition comprises a substantially layered perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph

the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 168 recites:

168. A method according to claim 133 wherein said superconducting oxide composition comprises a substantially **layered perovskite** crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 169 recites:

169. A method according to claim 134 wherein said transistor metal oxide comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot

practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

#### Claim 170 recites:

170. A method according to claim 135 wherein said copper oxide comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 171 recites:

171. A method according to claim 136 wherein said composition comprises a **substantially layered** perovskite crystal structure.

## Claim 172 recites:

172. A method according to claim 137 wherein said composition of matter comprises a **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 173 recites:

173. A method according to claim 138 wherein said composition of matter comprises **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 174 recites:

174. A method according to claim 139 wherein said composition of matter comprises a **substantially layered** perovskite crystal structure.

# Claim 175 recites:

175. A method according to claim 140 wherein said composition of matter comprises **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

## Claim 176 recites

176. A method according to claim 141 wherein said transistor metal oxide comprises **substantially layered** perovskite crystal structure.

As the rejections under §102(a) and §103(a) ASA provides no TSMA for the limitation highlighted in the claim language above. As to enablement under §112, first paragraph

the Examiner has provided no extrinsic evidence that a person of skill in the art cannot practice the invention of this claim from applicants teaching and from what is know to a person of skill in the art.

# Claim 177 recites:

177. A method according to claim 142 wherein said copper oxide composition comprises **substantially layered** perovskite crystal structure

## CONCLUSION

In view of the argument herein, Applicants request the Board:

- 1. reverse the determination that claims 24-26, 86-90, 96-135 and 137-177 are not supported by the priority document;
- 2. reverse the rejection of claims 24-26, 86-90, 96-135 and 137-177 as anticipated under 35 USC 102(a) by the Asahi Shinbum article;
- 3. reverse the rejection of claims 24-26, 86-90, 96-135 and 137-177 as obvious under 35 USC 103(a) in view of the Asahi Shinbum article;
- 4. reverse the rejection of claims 24-26, 86-90, 96-135 and 137-142 as not enabled under 35 USC 112, first paragraph; and
- 5. reverse the rejection of claims 86-87, 96-108, 115, 118, 120, 122, 123, 124,129-135 and 137-142 indefinite under 35 USC 112 second paragraph.

Please charge any fee necessary to enter this paper to deposit account 09-0468.

Respectfully submitted,

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# APPENDIX CLAIMS

24. A method including the steps of forming a transition metal oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K,

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase, and

passing an electrical supercurrent through said transition metal oxide while it is in said superconducting state.

- 25. The method of claim 24, where said transition metal oxide is comprised of a transition metal capable of exhibiting multivalent states.
- 26. The method of claim 24, where said transition metal oxide is comprised of a Cu oxide.
- 86. A method, comprising the steps of:

forming a composition including a transition metal, a rare earth or rare earth-like element, an alkaline earth element, and oxygen, where said composition is a mixed

transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K,

maintaining said composition in said superconducting state at a temperature greater than 26°K, and

passing an electrical current through said composition while said composition is in said superconducting state.

- 87. The method of claim 86, where said transition metal is copper.
- 88. A method, including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K, maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state, and

passing an electrical current through said composition while said composition is in said superconductive state.

- 89. The method of claim 88, where said composition is comprised of a metal oxide.
- 90. The metal of claim 88, where said composition is comprised of a transition metal oxide.

- 96. A superconductive method for causing electric current flow in a superconductive state at a temperature in excess of 26 K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductor transition temperature  $T_c$  of greater than 26 K;
- (b) maintaining the superconductor element at a temperature above 26 K and below the superconductor transition temperature  $T_{\text{c}}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 97. The superconductive method according to claim 96 in which the copper-oxide compound of the superconductive composition includes at least one rare-earth or rare-earth-like element and at least one alkaline-earth element.
- 98. The superconductive method according to claim 97 in which the rare-earth or rare-earth-like element is lanthanum.
- 99. The superconductive method according to claim 97 in which the alkaline-earth element is barium.

- 100. The superconductive method according to claim 96 in which the copper-oxide compound of the superconductive composition includes mixed valent copper ions.
- 101. The superconductive method according to claim 100 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.
- 102. The superconductive method according to claim 101 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.
- 103. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one rare-earth or rare-earth-like element and at least one alkaline-earth element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than 26 K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 104. The superconductive method according to claim 103 in which the rare-earth or rare-earth-like element is lanthanum.
- 105. The superconductive method according to claim 103 in which the alkaline-earth element is barium.
- 106. The superconductive method according to claim 103 in which the copper-oxide compound of the superconductive composition includes mixed valent copper ions.
- 107. The superconductive method according to claim 106 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.
- 108. The superconductive method according to claim 107 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.
- 109. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element.

110. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

111. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and an element selected from the group consisting of Group II A element, a rare earth element and a Group III B element.

- 112. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, a rare earth element; and a Group III B element;

- (b) maintaining the superconductor element at a temperature above 26  $^{\circ}$ K and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 113. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^o$ K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and

- (c) causing an electric current to flow in the superconductor element.
- 114. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26 °K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

115. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26 °K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

116. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26 °K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A and at least one element selected from the group consisting of a rare earth element and a Group III B element.

117. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

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- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}\text{K}$  and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 118. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition

defining a superconductive-resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^{\circ}$ K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 119. A method including the steps of forming a transition metal oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said transitional metal oxide includes at least one element selected from the group consisting of a Group II A element and at lest one element selected from the group consisting of a rare earth element and a Group III B element.

120. A method comprising the steps of:

forming a composition including a transition metal, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, where said composition is a mixed transitional metal oxide formed from said transition metal and said oxygen, said mixed transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

121. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26 °K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a transitional metal oxide and at least one element selected from the group consisting of Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element.

122. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:

(a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;

- (b) maintaining the superconductor element at a temperature above 26  $^{\circ}$ K and below the superconductor transition  $T_{\rm c}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 123. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal-oxide compound having a layer-type perovskite-like crystal structure, the transition metal-oxide compound including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T p=0, the transition-onset temperature T being greater than 26°K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and

- (c) causing an electric current to flow in the superconductor element.
- 124. A method including the steps of forming copper oxide having a phase therein which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said material at a temperature less than said critical temperature to produce said superconducting state in said phase;

passing an electrical supercurrent through said copper oxide while it is in said superconducting state;

said copper oxide includes at least one element selected from group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

125. A method comprising the steps of:

forming a composition including copper, oxygen and an element selected from the group consisting of at least one Group II A element and at least one element selected from the group consisting of a rare earth element at least one element selected from the group consisting of a Group III B element, where said composition is a mixed

copper oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K;

maintaining said composition in said superconducting state at a temperature greater than 26°K; and

passing an electrical current through said composition while said composition is in said superconducting state.

126. A method including the steps of:

forming a composition exhibiting a superconductive state at a temperature in excess of 26°K;

maintaining said composition at a temperature in excess of 26°K at which temperature said composition exhibits said superconductive state;

passing an electrical current through said composition while said composition is in said superconductive state; and

said composition including a copper oxide and at least one element selected from the group consisting of Group II A element, at least one element selected from the group

consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element.

- 127. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting

essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element;

- (b) maintaining the superconductor element at a temperature above 26  $^{\circ}$ K and below the superconductor transition temperature  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 128. A superconductive method for conducting an electric current essentially without resistive losses, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a layer-type perovskite-like crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a group II A element, at least one element selected from the group consisting of a rare earth element and at least one element selected from the group consisting of a Group III B element, the composition having a superconductive-resistive transition temperature defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T<sub>p=0</sub>, the transition-onset temperature T<sub>c</sub> being greater than 26°K;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 129. A method comprising providing a composition having a transition temperature greater than 26°K, the composition including a rare earth or alkaline earth element, a transition metal element capable of exhibiting multivalent states and oxygen, including at least one phase that exhibits superconductivity at temperature in excess of 26°K,

maintaining said composition at said temperature to exhibit said superconductivity and passing an electrical superconducting current through said composition with said phrase exhibiting said superconductivity.

- 130. A method comprising providing a superconducting transition metal oxide having a superconductive onset temperature greater than 26°K, maintaining said superconducting transition metal oxide at a temperature less than said superconducting onset temperature and flowing a superconducting current therein.
- 131. A method comprising providing a superconducting copper oxide having a superconductive onset temperature greater than 26°K, maintaining said superconducting copper oxide at a temperature less than said superconducting onset temperature and flowing a superconducting current in said superconducting oxide.
- 132. A method comprising providing a superconducting oxide composition having a superconductive onset temperature greater than 26°K, maintaining said superconducting copper oxide at a temperature less than said superconducting onset temperature and flowing a superconducting current therein, said composition comprising at least one each of rare earth, an alkaline earth, and copper.
- 133. A method comprising providing a superconducting oxide composition having a superconductive onset temperature greater than 26°K, maintaining said

superconducting copper oxide at a temperature less than said superconducting onset temperature and flowing a superconducting electrical current therein, said composition comprising at least one each of a Group III B element, an alkaline earth, and copper.

134. A method comprising flowing a superconducting electrical current in a transition metal oxide having a  $T_c$  greater than 26°K and maintaining said transition metal oxide at a temperature less than said  $T_c$ .

135. A method comprising flowing a superconducting current in a copper oxide having a  $T_c$  greater than 26°K and maintaining said copper oxide at a temperature less than said  $T_c$ .

136. A method comprising the steps of:

forming a composition of the formula Ba<sub>x</sub>La<sub>x-5</sub>, Cu<sub>5</sub>O<sub>Y</sub>, wherein x is from about 0.75 to about 1 and y is the oxygen deficiency resulting from annealing said composition at temperatures from about 540°C to about 950°C and for times of about 15 minutes to about 12 hours, said composition having a metal oxide phase which exhibits a superconducting state at a critical temperature in excess of 26°K;

maintaining the temperature of said composition at a temperature less than said critical temperature to induce said superconducting state in said metal oxide phase; and

passing an electrical current through said composition while said metal oxide phase is in said superconducting state.

- 137. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K, said composition comprising at least one each of a III B element, an alkaline earth, and copper oxide and maintaining said composition of matter at a temperature less than  $T_c$ .
- 138. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K, said composition comprising at least one each of a rare earth, alkaline earth, and copper oxide and maintaining said composition of matter at a temperature less than said  $T_c$ .
- 139. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K, said composition comprising at least one each of a rare earth, and copper oxide and maintaining said composition of matter at a temperature less than said  $T_c$ .
- 140. A method comprising flowing a superconducting electrical current in a composition of matter having a  $T_c$  greater than 26°K carrying, said composition comprising at least one each of a III B element, and copper oxide and maintaining said composition of matter at a temperature less than said  $T_c$ .

- 141. A method comprising flowing a superconducting electrical current in a transition metal oxide comprising a  $T_c>26^\circ K$  and maintaining said transition metal oxide at a temperature less than said  $T_c$ .
- 142. A method comprising flowing a superconducting electrical current in a copper oxide composition of matter comprising a T<sub>c</sub>>26°K and maintaining said copper oxide composition of matter at a temperature less than said T<sub>c</sub>.
- 143. A method, comprising the steps of:

forming a composition including a transition metal, a group IIIB element, an alkaline earth element, and oxygen, where said composition is a mixed transition metal oxide having a non-stoichiometric amount of oxygen therein and exhibiting a superconducting state at a temperature greater than 26°K,

maintaining said composition in said superconducting state at a temperature greater than 26°K, and

passing an electrical current through said composition while said composition is in said superconducting state.

144. The method of claim 143, where said transition metal is copper.

- 145. A superconductive method for causing electric current flow in a superconductive state at a temperature in excess of 26 K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductor transition temperature  $T_c$  of greater than 26 K;
- b) maintaining the superconductor element at a temperature above 26 K and below the superconductor transition temperature  $T_{\rm c}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 146. The superconductive method according to claim 145 in which the copper-oxide compound of the superconductive composition includes at least one element selected from the group consisting of a rare-earth element and a Group III B element and at least one alkaline-earth element.
- 147. The superconductive method according to claim 146 in which the rare-earth or rare-earth-like element is lanthanum.
- 148. The superconductive method according to claim 146 in which the alkaline-earth element is barium.

- 149. The superconductive method according to claim 145 in which the copper-oxide compound of the superconductive composition includes mixed valent copper ions.
- 150. The superconductive method according to claim 149 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.
- 151. The superconductive method according to claim 150 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.
- 152. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a rare-earth element and a Group III B element and at least one alkaline-earth element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than 26 K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $\mathsf{T}_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 153. The superconductive method according to claim 152 in which said at least one element is lanthanum.
- 154. The superconductive method according to claim 152 in which the alkaline-earth element is barium.
- 155. The superconductive method according to claim 152 in which the copper-oxide compound of the superconductive composition includes mixed valent copper ions.
- 156. The superconductive method according to claim 155 in which the copper-oxide compound includes at least one element in a nonstoichiometric atomic proportion.
- 157. The superconductive method according to claim 156 in which oxygen is present in the copper-oxide compound in a nonstoichiometric atomic proportion.
- 158. A superconductive method for causing electric-current flow

in a superconductive state at a temperature in excess of 26 °K, comprising:

- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element, a rare earth element; and a Group III B element;
- (b) maintaining the superconductor element at a temperature above 26  $^{\circ}$ K and below the superconductor transition temperature  $T_{\circ}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 159. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a Group II A element, a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition

temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than 26°K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 160. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above 26  $^{\circ}$ K and below the superconductor transition temperature  $T_{\circ}$  of the superconductive composition; and

- (c) causing an electric current to flow in the superconductor element.
- 161. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a copper-oxide compound having a substantially layered perovskite crystal structure, the copper-oxide compound including at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive-resistive-transition temperature range between an upper limit defined by a transition-onset temperature  $T_c$  and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$ , the transition-onset temperature  $T_c$  being greater than  $26^{\circ}K$ ;
- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.

- 162. A superconductive method for causing electric-current flow in a superconductive state at a temperature in excess of 26°K, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal oxide compound having a substantially layered perovskite crystal structure, the composition having a superconductive transition temperature T<sub>c</sub> of greater than 26°K, said superconductive composition includes at least one element selected from the group consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element;
- (b) maintaining the superconductor element at a temperature above  $26\,^{\circ}$ K and below the superconductor transition  $T_c$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 163. A superconductive method for conducting an electric current essentially without resistive losses, comprising:
- (a) providing a superconductor element made of a superconductive composition, the superconductive composition consisting essentially of a transition metal-oxide compound having a substantially layered perovskite crystal structure, the transition metal-oxide compound including at least one element selected from the group

consisting of a Group II A element and at least one element selected from the group consisting of a rare earth element and a Group III B element, the composition having a superconductive/resistive transition defining a superconductive/resistive-transition temperature range between an upper limit defined by a transition-onset temperature T<sub>c</sub> and a lower limit defined by an effectively-zero-bulk-resistivity intercept temperature T<sub>p=0</sub>, the transition-onset temperature T<sub>c</sub> being greater than 26°K;

- (b) maintaining the superconductor element at a temperature below the effectively-zero-bulk-resistivity intercept temperature  $T_{p=0}$  of the superconductive composition; and
- (c) causing an electric current to flow in the superconductor element.
- 164. A method according to claim 129 wherein said composition comprises a substantially layered perovskite crystal structure.
- 165. A method according to claim 130 wherein said superconducting transistor metal oxide comprises a substantially layered perovskite crystal structure.
- 166. A method according to claim 131 wherein said superconducting copper oxide comprises a substantially layered perovskite crystal structure.

- 167. A method according to claim 132 wherein said superconducting oxide composition comprises a substantially layered perovskite crystal structure.
- 168. A method according to claim 133 wherein said superconducting oxide composition comprises a substantially layered perovskite crystal structure.
- 169. A method according to claim 134 wherein said transistor metal oxide comprises a substantially layered perovskite crystal structure.
- 170. A method according to claim 135 wherein said copper oxide comprises a substantially layered perovskite crystal structure.
- 171. A method according to claim 136 wherein said composition comprises a substantially layered perovskite crystal structure.
- 172. A method according to claim 137 wherein said composition of matter comprises a substantially layered perovskite crystal structure.
- 173. A method according to claim 138 wherein said composition of matter comprises substantially layered perovskite crystal structure.
- 174. A method according to claim 139 wherein said composition of matter comprises a substantially layered perovskite crystal structure.

- 175. A method according to claim 140 wherein said composition of matter comprises substantially layered perovskite crystal structure.
- 176. A method according to claim 141 wherein said transistor metal oxide comprises substantially layered perovskite crystal structure.
- 177. A method according to claim 142 wherein said copper oxide composition comprises substantially layered perovskite crystal structure.

## ATTACHMENT A

# Inorganic Chemistry

AN ADVANCED TEXTBOOK

### THERALD MOELLER

Associate Professor of Chemistry University of Illinois

New York · JOHN WILEY & SONS, Inc. London · CHAPMAN & HALL, Limited

Attachment A pager

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ric compounds as opposed to the normal Daltonide or stoichiometric compounds. As examples, one may cite certain metallic hydrides such as VH<sub>0.56</sub>, CeH<sub>2.69</sub> (p. 411); certain oxides such as TiO<sub>1.7-1.8</sub>, FeO<sub>1.055</sub>, WO<sub>2.88-2.92</sub>; such sulfides, selenides, and tellurides as Cu<sub>1.7</sub>S, Cu<sub>1.6</sub>Se, Cu<sub>1.6</sub>Te, CuFeS<sub>1.94</sub>; the tungsten bronzes, Na<sub>x</sub>WO<sub>3</sub>; etc. Combinations of these types are particularly common among minerals.

Lack of true stoichiometry of this type is associated with so-called defect crystal lattices. Defects in a crystal lattice amount to variations from the regularity which characterizes the material as a whole. They are of two types:

1. Frenkel defects, in which certain atoms or ions have migrated to interstitial positions some distance removed from the "holes" which they vacated.

2. Schottky defects, in which "holes" are left in random fashion throughout the crystal because of migration of atoms or ions to the surface of the material.

Although both types of defect probably characterize crystals of non-stoichiometric compounds, the Schottky defects are the more important. Obviously detectable departure from true stoichiometric composition can result only if serious defects are present. It would follow, therefore, that many apparently stoichiometric compounds are not truly so. If excess metal is present in a crystal, it may also result from partial reduction of high-valent cations; whereas if excess non-metal is present, higher valent cations or lower valent anions than those normally present may be responsible. Many instances are known of multiple oxidation number in a single crystal. Non-stoichiometric compounds often show semi-conductivity, fluorescence, and centers of color. For a comprehensive discussion of this rather complex subject, a detailed review should be consulted.

## SUMMARY OF BOND TYPES

The important linkages which hold together the components of crystalline solids and their general characteristics may be summarized as follows:

1. Ionic linkages, in which the crystals are made up of regular geometrical arrangements of positive and negative ions. Such solids tend to possess high melting and boiling points, are hard and difficult to deform, and tend to be soluble in polar solvents. When dissolved in such solvents or fused, they are excellent conductors. Crystals

90 J. S. Anderson: Ann. Reports, 43, 104 (1946).

A Hachment A page 3

overcome. Such cage compounds have been called clathrate compounds89 (Latin clathratus, enclosed by cross bars of a grating). In general, they occur when mixtures of the components are crystallized under optimum conditions. Their properties are roughly those of the enclosing material. Such compounds are stable at ordinary temperatures with respect to decomposition into their components, but melting or dissolution permits the enclosed component to escape. Examples are hydroquinone compounds which approach the composition (C<sub>6</sub>H<sub>6</sub>O<sub>2</sub>)<sub>2</sub>·X(X = HCl, HBr, H<sub>2</sub>S, CH<sub>3</sub>OH, SO<sub>2</sub>, CO<sub>2</sub>, HCN, etc.); amine compounds containing sulfurous acid, e.g. (p-H2NC6H4- $NH_2$ ) 9·  $H_2SO_3$ ; phenol compounds, e.g.  $(C_6H_6O)_4\cdot SO_2$ ,  $(C_6H_6O)_5\cdot SO_2$ , (C<sub>6</sub>H<sub>6</sub>O)<sub>8</sub>·CO<sub>2</sub>; and certain compounds of the inert gas elements (pp. 382-383).

It is obvious that the conditions under which clathrate compounds can form are limited and highly specific. Among those of importance

are:

1. An open crystal structure in the enclosing component. necessitates directed linkages holding the molecule and crystal together, sufficient extension of the groups to form a cavity of suitable size, and a rigid structure.

2. Small access holes to the enclosed cavity. This may result from either proper disposition of groups in the formation of the crystal or

sufficient surface area in the enclosing groups.

3. Ready availability of the trapped component at the time when the cavity is closed.

Such compounds are of considerable theoretical interest but are lacking in practical importance. Information on possible arrangements in clathrate compounds and the structures which lead to them is to be found in Powell's discussions.89

# NON-STOICHIOMETRIC COMPOUNDS

The law of definite proportions is one of the basic tenets of chemistry Its validity is indicated by the restrictions imposed upon bond forms tion where electrons are involved as already outlined, and its application is generally the assumed basis for any type of chemical combination tion. There are, however, many instances of apparent departure from this rule among solid compounds. Such compounds do not possess the exact compositions which are predicted from electronic considerations alone and are commonly referred to as Berthollide or non-stoichiomet

89 H. M. Powell: J. Chem. Soc., 1948, 61; Endeavour, 9, 154 (1950); Research, 1 353 (1947-1948).

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Department of Chemistry
The City College of the
City University of New York



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matter: the mass of a chemically reacting system remains constant. This law is consistent with the data obtained with the most precise balances available. If matter is created or destroyed, the quantity is less than can be detected with the best available balance.

## 4.2 THE LAW OF DEFINITE PROPORTIONS

Analyses of compounds show that when elements form a given compound, they always combine in the same ratio by weight. For example, independently of the source or method of formation, silicon dioxide, SiO<sub>2</sub>, contains 46.7% by weight of silicon and 53.3% of oxygen. This knowledge is summarized in the law of definite proportions: the weight composition of a given compound is constant.

EXAMPLE 1 10.0 g of silicon dust, Si, is exploded with 100.0 g of oxygen, O<sub>2</sub>, forming silicon dioxide, SiO<sub>2</sub>. How many grams of SiO<sub>2</sub> are formed and how many grams of O<sub>2</sub> remain uncombined?

ANSWER Since 46.7 g of Si combines with 53.3 g of O<sub>2</sub>, the quantity of O<sub>2</sub> required per gram of Si is

53.3 g O<sub>2</sub> 46.7 g Si

and, therefore, for 10.0 g of Si, the quantity of O2 required is

$$10.0 \text{ g/Si} \frac{53.3 \text{ g/O}_2}{46.7 \text{ g/Si}} = 11.4 \text{ g/O}_2$$

Hence, the weight of SiO<sub>2</sub> formed is 10.0 g + 11.4 g = 21.4 g and the weight of uncombined O<sub>2</sub> is 100.0 g - 11.4 g = 88.6 g.

### 4.3 THE ATOMIC THEORY

The weight relationships of substances participating in chemical reactions are clearly explained in terms of the atomic theory. Although John Dalton (1803) is generally recognized as the inventor of the theory, he was anticipated by other scientists, particularly William Higgins (1789). Thus, it appears that the law of multiple proportions (Section 4.4) was foreshadowed by Higgins and Dalton from their respective atomic theories. A verified prediction made by a theory constitutes the strongest argument in its favor. However, the novel and central point of Dalton's activities was the attempt to determine the relative weights of atoms. This goal focused attention upon the theory, and revealed a new field of human endeavor that ultimately made chemistry a systematized body of knowledge.

The assumptions of the atomic theory were

- (i) The elements are composed of indivisible particles called atoms.
- (ii) All the atoms of a given element possess identical properties, for example, mass.

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# TRANSPORT PROPERTIES

#### A. INTRODUCTION

The principal applications of superconductors are based upon their ability to carry electric current without any loss, and therefore it is important to understand their transport properties. This chapter begins with a discussion of resistivity and critical current flow in the absence of externally applied fields. This is followed by a discussion of several techniques involving applied fields and thermal effects. The chapter concludes with sections on tunneling and the Josephson effect.

#### **B. CURRENT FLOW**

Electric currents that flow through a superconductor owing to the action of an external source of potential are called transport currents, and those that arise in an external magnetic field to cancel the magnetic flux inside the superconductor are called diamagnetic screening currents or shielding currents. In magnet applications transport currents are started by an external source and continue to flow (persist) after its removal, while in long-distance electrical transmission applications the source voltage continues to be applied.

Current densities J are intrinsic properties and hence are more useful than currents I for quantitative comparisons between different superconductors. Transport current densities can be comparable in magnitude to shielding current densities.

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The velocity of electrons at the Fermi surface  $v_F$  was estimated to be  $10^7$  cm/sec in these materials, which is of the same order as in A-15 compounds, and  $\frac{1}{10}$  of the value in aluminum (Garoc).

#### 1. Resistivity

A susceptibility measurement is a better thermodynamic indicator of the superconducting state because magnetization is a thermodynamic state variable. The resistivity, on the other hand, is easier to measure, and can be a better guide for applications. The temperature of zero resistivity shows when continuous superconducting paths are in place between the electrodes. Filimentary paths can produce sharp drops in resistivity at higher temperatures than pronounced onsets of diamagnetism. This can be described in terms of two- and three-dimensional percolation thresholds (see Section III-E).

Many investigators have published figures of resistivity or resistance versus temperature, since this is the most popular way to determine the critical temperature and the sharpness of the transition. It should be remembered that if the specimen is porous, accurate determinations of the resistivity cannot readily be made because of the presence of voids and intergrain problems. In almost all cases  $T_{\rm c}$  determined from the resistivity midpoint is at a higher temperature than its susceptibility counterpart.

Good conductors such as copper and silver have room-temperature resistivities of about 1.5  $\mu\Omega$  cm, and at liquid nitrogen temperature the resistivity typically decreases by a factor of 6-8, as shown by the data in Table X-1. The elemental superconductors, such as Nd, Pb, and Sn, have room-temperature resistivities a factor of 10 greater than good conductors. The other metallic elements present in oxide superconductors, namely, Ba, Bi, Ca, La, Sr, Tl, and Y, have resistivities 10-80 times that of Cu. The copper oxide superconductors have even higher room-temperature resistivities, over three orders of magnitude greater than that of metallic copper, which puts them within a factor of 3 or 4 of the semiconductor range, as shown by the data in Table X-1. The resistivity of these materials above  $T_c$  decreases more or less linearly with decreasing temperature (cf. Fig. VII-11) down to the neighborhood of  $T_c$ , with a drop by a factor of 2 or 3 from room temperature to this point, as shown by the data in the table.

Needless to say, the concept of resistivity is not a meaningful one to apply to a superconductor below  $T_c$ . Nevertheless, it is instructive to study the low-temperature resistance in nonsuperconducting compounds that are closely related to superconductors. For example, in nonsuperconducting crystals of  $(La_{1-x}-Sr_x)_2(Cu_{1-y}Li_y)O_{4-\delta}$  a variable range type of hopping resistance, that is,  $R \approx \exp[(T_0/T)^4]$ , was reported. From this it has been argued that the poor conductivity is not due to a large gap, but rather to localization of the states at  $E_F$  (Kastn).

The resistivity in the high-temperature ( $80 \le T \le 1200 \text{ K}$ ) region is linear with the temperature for T < 600 K and superlinear above 600 K. This linearity has been linked to the two-dimensional character of the electron transport

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TABLE X-1. Examples of Resistivities at Room Temperature  $\rho_{300}$ , at Low Temperature  $\rho(T)$ , and Their Ratio  $\rho_{300}/\rho(T)^a$ 

Material	ρ <sub>300</sub> (μΩ cm)	$ ho(T)$ ( $\mu\Omega$ cm)	<i>T</i> (K)	$\rho_{300}/\rho(T)$	Rei
Cu	1.68	0.18	77	9.3	
Ag	1.60	0.26	77	6.2	
Pt	10.6	1.74	77	6.1	
Sn	12.4		• •	0.1	
Nb	12.5				
TI	18				
Pb	20				
Sr	23				
Ca	53				
Y	57				
La	58				
Ba	60				
Bi	119				
LaSr*	2200	430	44	5.1	77 .
	2700	850	50	3.1	Koba
	2300	510	50	3.2 4.5	Penne
LaSr (0.05)		950(  )	40	4.3	Taral
		19000(⊥)	40		Hidak
(0.1)		_,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	50	3.6	Hidal
		1600	44	3.0	Coppe
	1200		• • •		Koba
.aBa(0.1)			50	4.0	Tonou
′Ba∗	2000	1000	<b>9</b> 5	2.0	Coppe
	650	225	95	2.9	Bonn
	900	470	100	1.9	Caval
	4000	2000	100	2.0	Mawd
		730	120	1.9	Neum
	1350	680	100	2.0	Panso
( <b>µ</b>  )	450	200	100	2.3	Penne
(AT)	13000	18000	100	0.7	Tozer
<sub>0.6</sub> Ba <sub>0.4</sub> CuO <sub>3</sub>	$1.5 \times 10^{6}$		100	0.7	Tozer
yBa <b>∗</b>				~1.3	Tonou
uBa <b>∗</b>	720	410	100	1.8	Mapil
mBa∗	4600	1900	100	2.4	Hikit
bBa≰		<del>-</del>		~3.3	Neume Mapil

<sup>&</sup>lt;sup>a</sup>Typical semiconductors have values from  $10^4$  to  $10^{15}$   $\mu\Omega$  cm and insulators range from  $10^{20}$  to  $10^{28}$   $\mu\Omega$  cm. The notation  $\parallel$  and  $\perp$  refers to resistivity measurements made parallel to and perpendicular to the Cu–O planes, respectively.

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ange from 10<sup>20</sup> to 10<sup>28</sup> I to and perpendicular

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(Micna). Applying the Mott-Ioffe-Regel rule (minimum scattering length < mean free path) to the observed normal state resistivity gave electron-phonon couplings in both LaSr\* ( $\lambda = 0.1$ ,  $\lambda_{max} = 0.45$ ) and YBa\* ( $\lambda = 0.3$ ,  $\lambda_{max} = 1.2$ ) which were too small to account for the observed  $T_c$  (Gurv1, Gurv2).

An exponential dependence of the resistivity of YBa\* on the temperature was observed between 80 and 1240 K (Fishe). This may occur via tunneling of electrons through barriers. The value of the exponent was different above and below the temperature  $T_*$  (700-750), which is near the tetragonal-to-orthorhombic transition. The temperature dependence of the resistivity appears to result from the loss of oxygen during heating, and the following expression was proposed to reflect this dependence:

$$\rho(T) = \frac{AT}{1 - \delta(T)} \tag{X-1}$$

where A is temperature insensitive and  $\delta(T)$  is the oxygen content factor in the formula YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Chaki; see also Fiory). A break in the slope of the logarithmic derivative  $(T/\rho)d\rho/dT$  plotted against T occurs at the orthorhombic-to-tetragonal phase transition (Fiory). Evidence for an n-to-p type transition was also reported (Choi1).

The resistivity is much higher when measured perpendicular to the ab planes (i.e., along the c axis) than it is parallel to the planes. Measured ratios  $\rho_{\perp}/\rho_{\parallel}$  are about 20 for LaSrCuO (Hidak), 50 for YBaCuO (Tozer), and  $10^5$  for BiSrCaCuO. We see from Table X-1 that typical measured resistivities, which are on polycrystalline specimens, are much closer to the in-plane values.

Hysteresis effects have been seen in the resistance versus temperature curves, as illustrated on Fig. X-1 for  $(Y_{0.875}Ba_{0.125})_2CuO_{4-\delta}$  (Taras). The 2-K shift in  $T_c$  for decreasing and increasing temperature measurements is about half of the width of the transition.

## 2. Critical Current Density

When the current density in a superconductor exceeds a value called the critical current density  $J_C$ , the superconductivity is destroyed. This is called the Silsbee effect. The value of  $J_C(T)$  increases from zero at  $T = T_c$  to a maximum value  $J_C(O)$  at 0 K (Leide). Figure X-2 shows the magnetic field dependence of  $J_C$  for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> from 0.5 to 6 T for several temperatures in the range from 4.2 to 83 K (e.g., Panso; see also Ekinz, Jones).

The value of  $J_C$  can be determined directly by the resistivity method by measuring the current at which a small voltage (typically 1  $\mu$ V) is induced across the sample ( $\approx$ 1 cm) in a four-probe resistivity arrangement. An indirect method uses a magnetization versus field hysteresis loop through the expression (Kumak, see also Sunzz, Xiao2).

$$J_{\rm C} = 30 \ \Delta M/d \qquad \text{A/cm}^2 \tag{X-2}$$

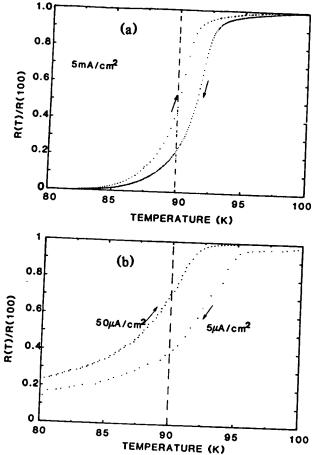


Fig. X-1. Hysteresis of resistance versus temperature for two samples of YBaCuO, for increasing and decreasing temperature, as indicated by the arrows. (a) current density 5 mA/cm<sup>2</sup>; (b) current density 50  $\mu$ A/cm<sup>2</sup> for increasing temperature and 5  $\mu$ A/cm<sup>2</sup> for decreasing temperature (Taras).

where  $\Delta M$  is the hysteresis of magnetization per unit volume in electromagnetic units per cubic centimeter and d is the size of the sample in centimeters.

The  $J_C$  values measured directly are called transport currents and those determined from hysteresis loops are called magnetization currents. Transport currents were found to be smaller than magnetization currents in the LaSrCuO (e.g., Larba) and YBaCuO (e.g., Kuma1, Togan) systems. This could be caused by granularity and intergrain contact, and improving sample quality might bring transport currents closer to their magnetization current counterparts (Wuhlz). In contrast to this, magnetization and transport critical currents of YBa\* epitaxial films were reported to be the same (Chaud, Ohzzz).

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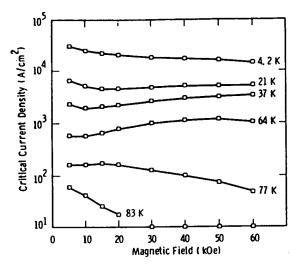


Fig. X-2. Magnetic field dependence of critical current densities of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> obtained from hysteresis loops (Panso) (10 kG = 1 T).

Critical current densities for a number of LaSrCuO and YBaCuO materials are listed in Table X-2 (e.g., Camp1, Chau2, Chau3, Daizz, Jinz2, Kagos, Umeza, Xiao2). We see from the table that the YBaCuO compounds tend to have higher values of  $J_C$  than LaSrCuO. For the same material  $J_C$  is considerably larger at 4.2 K than at 77 K. It is also quite anisotropic, with critical current densities much larger parallel to the Cu-O planes than perpendicular to them. Jin et al. (Palca) employed a technique called melt-textured growth to achieve  $J_C$  as high as  $10^3$  A/cm² in a magnetic field of 1 T at 77 K. Values of  $J_C = 10^6$  A/cm² and greater have been reported in epitaxial thin films (Chaud, Kwoz1, Ohzzz) and single crystals (Crabt, Ding1, Wort1).

Grinding and heat treating samples of LaSrCuO and YBaCuO was found to appreciably increase the critical current density at 4 K (Suena). High critical currents require efficient flux pinning since  $J_{\rm C}$  increases with the pinning force (Huebe, p. 125). Weak pinning leads to flux creep and low critical currents (e.g., Giova).

Very high values of  $J_{\rm C}$  are needed for magnet materials. A niobium-titanium filament has been reported with  $J_{\rm C}$  values as high as 3.7  $\times$  10<sup>5</sup> A/cm<sup>2</sup> at 5 T (Cheng). The Superconducting Super Collider and Relativisitic Heavy Ion Collider accelerators require 5- $\mu$ m wire filaments that support  $J_{\rm C}$  of at least 2.8  $\times$  10<sup>5</sup> A/cm<sup>2</sup> in a 5-T magnetic field (Grego). At present  $J_{\rm C}$  values of technologically suitable oxide materials at 77 K are too small for such high-field magnet applications. Although the oxide superconductors do not yet compete with the old ones in critical currents, they are superior in their critical field ( $H_{\rm C2}$ ) capability, as shown in Fig. I-2.

s of YBaCuO, for a) current density and 5 μA/cm<sup>2</sup> for

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TABLE X-2. Critical Current Densities

	carrent Densines					
Compound	$J_c$ (A/cm <sup>2</sup> )	7 (K)	H (T)	Measurement		
LaSr*	105			DOIDER	Comments	Ref.
LaSr(0.05)	. 2	4.2	0 035	Mg		Suena
	-	4.2	10 <sup>-3</sup> -3	i f		Larba
:	0.75	4.2	5 6	<b>≒</b> €		Larba
Y Ba*	<b>&gt;10</b>	4.2		<u> </u>	ı	Larba
	> 105	11		Mg	E-film	Chaud
	10\$	72		Mg	E-film	Chaud
	$1.4 \times 10^{\circ}(\perp)$	်လ	0-1	7	E-film	Chau1
	$1.4 \times 10^4$	· v	, P	Mg.	M-xt	Crabt
	$1.1 \times 10^{\circ}(1)$	77	† <b>-</b>	Mg ;	P-xt	Crabt
	$4.3 \times 10^{3}(\perp)$	12	-	Mg	M-xt	Crapt
	$1.2 \times 10^{2}$	: £	7 0	Mg	M-xt	Crabt
	4 × 10°	, , 4 S	0.0	Mg	P-xt	Craht
	$3 \times 10^{6}(\perp)$	5.4 2.5		Mg	M-xt	Dino1
	×10°	5.4	7	Mg.	M-xt	Dingl
	1-200	0	,	Σ <del>[</del>	M-xt	Ding1
			•	11		Ekinz
		77	0	Ė	Je 1/Jell ~ 6	Glowa
	3 × 104	77	1	Ϋ́	Sintered rod	Jinzz
	10°	4.2		្នុះ	E-film, be plane	Kumak Kwoz1

Kwozl Larbi Larbi

E-film, bc plane

Tr Mg Xg X

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t 4.2 5.4 1

> 1.5-2 × 10<sup>4</sup> 235

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Ding1 Ding1 Ekinz	Glowa Jinzz Kumak Kwoz1
M-xt M-xt	J <sub>c1</sub> /J <sub>cl</sub> ~ 6 sintered rod P.xt E-film, bc plane
Mg Mg Tr	Tr Mg Tr
γ •	
0 4.S	77 77 4.2
>10° 1-200	620 3 × 10 <sup>4</sup> 10 <sup>6</sup>

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	<del>ار</del> ا	77		Ţ	E-film, be plane	Kwoz1
	1.5-2 × 104	4.2	1-10	Mg		Larb1
	235	77	:	Mg		Larb1
	€ €	77	· <b>v</b> c	Mg.		Larb1
	0 9-11 × 10 <sup>5</sup>	4.2	0.3	W W	E-film	Ohzzz
	1-5 × 10 <sup>4</sup>	78	0	Mg	E-film	Ohzzz
	0.4-4 × 10*	. 82	0.3	Mg	E-film	Ohzzz
	$1.5-3 \times 10^4$	4.2	0.1-6	Mg		Panso
	50-200	11	0.1-6	Mg		Panso
	$9.4 \times 10^{4}(1)$	S	0.4	Mg	sinter forged	Songz
	S.9 × 10 <sup>4</sup> (II)	S	0.4	Mg	sinter forged	Songz
	10	4		Mg		Suena
	8 × 10	4.2	0.2	Mg		Togan
	V 102	77	0.1	Mg		Togan
	3.2 × 106( 1.)	4.5	.0	Mg	M-xt	Wortl
	1.6 × 105(  )	4.5	0	W W	M-xt	Wort
VBa.Cu.O.	168	24	•	) <b>,</b>		Capol
YBarCurO.	>1100	12	0	T		Caval
V. Ba. CiO	14	14	0	Ţ		Iguch
VRaCuOx	2800	4.2		Tr	pulsed	Jones
YBaCuO	050	77		Ţ	pulsed	Jones

<sup>&</sup>quot;E-film, epitaxial film; Mg, magnetization; M-xt, monocrystal; P-xt, polycrystal; Tr, transport. Some measurements were made in the presence of an applied magnetic field H, and the notation ∥ and ⊥ refers to currents measured with the field applied parallel and perpendicular to the Cu-O planes, respectively.

#### 3. Persistent Currents

The zero-resistance property of a superconductor implies that an electrical current flowing in a closed path should persist indefinitely. Several investigators have examined this property and set lower limits on the lifetime of the current and upper limits on its associated resistivity. The lifetime of the persistent or superconducting current in a cylindrical LaSrCuO sample is in excess of  $3 \times 10^6$  sec or more than a month, corresponding to a resistivity of less than  $3 \times 10^{-17} \Omega$  cm (Wells). Current lifetimes in loops of low-temperature superconductors are much longer and suggest an effective resistivity of  $<10^{-23} \Omega$  cm (Chand), close to the value of  $7 \times 10^{-23} \Omega$  cm reported for YBaCuO (Kedve). Other reported minimum resistivity determinations are:  $<10^{-9} \Omega$  cm (Iguch),  $4 \times 10^{-16} \Omega$  cm for  $(Y_{0.6}Ba_{0.4})_2$ CuO<sub>4</sub> (Skoln),  $<10^{-16} \Omega$  cm in YBa<sub>4</sub> (Tjuka), and  $2 \times 10^{-18} \Omega$  cm (Yehzz). Similar results  $(10^{-24} \Omega$  cm) were obtained for TlBaCaCuO.

The ceramic superconductors are granular and the relaxation of trapped field and critical current loops may be characteristic of glassy structures (cf. Section VIII-D-5). Because of the granularity even small fields can penetrate the materials. This property has been utilized to nondestructively read the supercurrent (Macf1).

# C. MISCELLANEOUS TRANSPORT PROPERTIES

In this section we will discuss some transport properties that depend upon the application of electric or magnetic fields, and some that involve thermal effects. Various transport results of YBa\*, namely, thermal conductivity, thermopower, Hall constant, and resistivity, were found to be consistent with ordinary metallic behavior with a strong phonon interaction (Uher2). It was concluded that there is no evidence for exotic electronic behavior in YBa\*.

### 1. Magnetoresistance

A number of investigators have studied the resistance versus temperature behavior in low (Hikam), high (Kwokz, Mats1, Mura1, Uher4), and very high (Ouss1) magnetic fields.

Very high field  $(H \le 43 \text{ T})$  longitudinal and transverse orientation studies of YBa\* (Ouss1) show that the magnetoresistance  $\Delta \rho(T,H) = \rho(T,H) - \rho(T<0)$  may be decomposed into three contributions

$$\Delta \rho = \Delta \rho^{M} + \Delta \rho^{N} + \Delta \rho^{S} \tag{X-3}$$

where  $\Delta \rho^{S}$  (>0) is the increase in resistance when the superconducting fluctuations are suppressed by the field,  $\Delta \rho^{N}$  is the normal or Lorentz magnetoresistance, and  $\Delta \rho^{M}$  is an unidentified component that may be associated with mag-

netic ordering at 50 K, and critical field

One of th resistivity me bility determ ments in m YBaCuO system downward t susceptibilit increase in f

#### 2. Hall Effe

The Hall effers in the ne carriers are drop to zero

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Fig. X-3. To fields from (

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t an electrical curveral investigators ime of the current f the persistent or nexcess of  $3 \times 10^6$  s than  $3 \times 10^{-17} \Omega$  perconductors are cm (Chand), close e). Other reported ),  $4 \times 10^{-16} \Omega$  cm and  $2 \times 10^{-18} \Omega$  1BaCaCuO.

on of trapped field ctures (cf. Section netrate the materil the supercurrent

t depend upon the ve thermal effects. ity, thermopower, ordinary metallic ncluded that there

emperature behavi very high (Ouss1)

entation studies of  $(T,H) - \rho(T < 0)$ 

(X-3)

onducting fluctuaentz magnetoresissociated with magnetic ordering. No change in the superconductivity was observed when H=43 T at 50 K, and only  $\approx 50\%$  normal phase resistance was found at 77 K. The upper critical field  $H_{C2}$  was estimated as  $\approx 125$  T.

One of the problems with comparing transport and magnetic  $T_c$  data is that resistivity measurements are generally made in zero magnetic field and susceptibility determinations require the presence of a field. The resistivity measurements in magnetic fields (Kobay, Wuzzz) show that for the LaSrCuO and YBaCuO systems, respectively, the transition temperature broadens and shifts downward by perhaps 1 K/T (Ihar1, Nakao), as shown in Fig. X-3. In an ac susceptibility determination a downward shift of  $T_c$  by 2 K was reported for an increase in field amplitude from 3.1 to 31  $\mu$ T (Odazz).

#### 2. Hall Effect

The Hall effect provides information on the sign and the mobility of charge carriers in the normal state, and usually a positive sign indicates that the majority carriers are holes. In the superconducting state the Hall voltage is expected to drop to zero (Hundl, Zhan1).

In this experiment a magnetic field  $H_0$  is applied perpendicular to the direction of the current flow through the sample. The Lorentz force of the magnetic field on the moving charge carriers produces a charge separation which induces an electric field  $E_{\rm x}$  perpendicular to the current and magnetic field directions. The Hall coefficient  $R_{\rm H}$  is the ratio

$$R_{\rm H} = E_{\rm x}/JH_0 \tag{X-4}$$

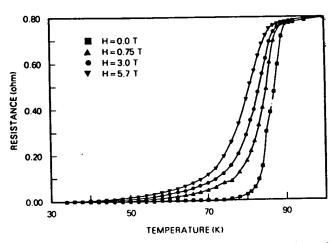


Fig. X-3. Temperature dependence of the resistance of YBaCuO in various magnetic fields from 0 to 5.7 T (Wuzzz).

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where J is the current density. When the charge carriers are electrons with the density n per cubic centimeter, the Hall coefficient is negative with the value

$$R_{\rm H} = -1/nec \tag{X-5}$$

in cgs units. A similar expression with a positive sign applies to hole conduction. Also of interest are the Hall mobility

$$\mu_{\rm H} = R_{\rm H}/\rho \tag{X-6}$$

where  $\rho$  is the resistivity, and the dimensionless Hall number  $V_0/R_{\rm H}e$ , where  $V_0$  is the volume per formula unit:

$$V_0 = 94 \text{ Å}^3 \quad \text{for } (\text{La}_{0.925}\text{Sr}_{0.075})_2\text{CuO}_4$$
 (X-7a)

$$V_0 = 174 \text{ Å}^3 \quad \text{for } YBa_2Cu_3O_7$$
 (X-7b)

Some authors find a strong temperature dependence of  $R_{\rm H}$  or the Hall number  $V_0/R_{\rm H}e$  for LaSrCuO (Tonou) and YBaCuO (Penne, Wangz). Others find a weak dependence for LaSrCuO (Hundl, Penne, Uchi1), and a large anomaly in  $R_{\rm H}$  near  $T_{\rm c}$  has been observed in YBaCuO (Gottw, Yongz). Figure X-4 shows  $R_{\rm H}$  of three substituted YBaCuO compounds increasing strongly with decreasing temperature (Cheon, Tana1). Figure X-5 shows the temperature dependence of the Hall mobility  $\mu_{\rm H}=R_{\rm H}/\rho$ , the Hall number  $V_0/R_{\rm H}e$  and the resistivity  $\rho$  of an epitaxial film of YBaCuO (Chau1, Penne). Hall-effect data on various compounds are listed in Table X-3.

Hall-effect data on LaSrCuO provided the room-temperature electron concentration  $n = 1.5 \times 10^{21}$ /cm<sup>3</sup> and the room-temperature mobility  $\mu_{\rm H} = 4.17$ 

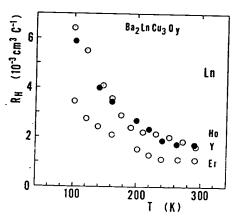


Fig. X-4. Temperature dependence of the Hall coefficient  $R_{\rm H}$  of LnBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> for Ln = Y, Ho, and Er (Tana1).

Fig. R<sub>H</sub>e,

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ctron con $u_H = 4.17$ 



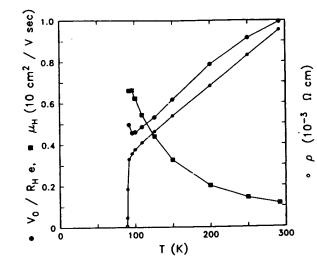


Fig. X-5. Transport data for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> showing the resistivity  $\rho$ , Hall number  $V_0/R_{\rm He}$ , and Hall mobility  $\mu_{\rm H}=R_{\rm H}/\rho$  versus temperature (Chau1).

cm<sup>2</sup>/V sec (Tonou). The electron concentration decreases with increasing temperature, probably owing to significant capture of thermally excited electrons at deep traps in grain boundaries surrounding the conductive channels. The Hall coefficient measurement of  $(La_{1-x}Sr_x)_2CuO_4$  single-crystal films grown epitaxially on SrTiO<sub>3</sub> provided a room-temperature carrier density  $n=6.8\times 10^{21}/cm^3$  which decreased notably with falling temperature (Suzuk).

Hall voltage measurements in LaSr\* indicated the presence of granular or inhomogeneous superconductivity (Hundl). The temperature dependence of the Hall coefficient of  $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_{4-\delta}$  was interpreted in terms of a relatively large phonon coupling and conduction via both electron and hole bands (Uher1, Uher3). The chemically determined electron deficiency or hole concentration in this compound exhibited a direct correlation with  $T_c$  for  $x \leq 0.15$ , suggestive of single-band transport and supportive of an all electronic mechanism for superconductivity (Shafe, Shaf1).

Hall-effect measurements on YBa<sub>2</sub>Cu<sub>3</sub>O<sub>8- $\delta$ </sub> single crystals with the magnetic field on  $\pm 1$  T in the ab plane yielded a negative Hall constant in the range -0.75 to  $-9 \times 10^{-10}$  m<sup>3</sup>/C corresponding to 1.2-1.5 electrons per formula unit (Tozer). A large temperature dependence in  $R_{\rm H}$  of YBa\* was attributed to multiband conduction (Hongm) and to the temperature dependence of the 2-D and 1-D chain mobility ratio (Wangz). The Hall coefficient was inversely dependent on the temperature in the compounds YBa\* and GdBa\*, which are judged as moderately heavy Fermion-like, with a Coulomb correlation energy comparable to or larger than the bandwidths (Cheon). Around the transition a larger increase was observed in  $R_{\rm H}$  for both YBa\* and DyBa\* (Yongz, Zhan1). In DyBa\* the peak in  $R_{\rm H}$  corresponds to  $n \approx 4 \times 10^{19}$  cm<sup>-3</sup>, and was interpreted as a grain

boundary boundary with decre change fro Several relaxation holelike in why sign (

### 3. Therm

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Figure: Seebeck co of x (Alle3 cient decre compound was observ result was cally to a v activated 1 range-hop

TABLE X-3. Hall Effect Data"

	Α.						,
Material	(cm <sup>3</sup> /°C)	$V_0/R_{ m H}e^b$	$^{n}$ (cm <sup>-3</sup> × 10 <sup>21</sup> )	$\mu$ (cm <sup>2</sup> /Vsec)	o (m)	T (3)	
LaSr*	+0.001				(he cui)	(¥)	Ket.
	+0.004		0.0			300	Hundi
		,	2.1	4.3	009	77	Onozz
1 2 5 7 (0 1)	300 0	0.3		+0.75	2600	300	Denne
Last(0.1)	200.0		1.5	-4.17	1200	300	Tonon
La-CuO	:		6.8	1.2	710	300	Curant
VRs.	1.0	•		1.0	10	300	Johin
*		1.0		+1.0	1000	300	Chant
		1.0 1.0		±0.5	1300	300	Penne
	0.0005-0.002	t.	•	₹0.8	950	300	Penne
			1.4			290	Wanga

"Hall coefficient  $R_{\rm H}$ , Hall number  $V_0/R_{\rm He}$ , carrier density n, mobility  $\mu$ , resistivity  $\rho$ , and measurement temperature T. The notation used is:  $(La_{1.2}Sr_{.2})_{.2}CuO_{4} = LaSr_{4}$ ,  $LaSr(0.075) = LaSr_{6}$ ,  $YBa_{2}Cu_{3}O_{7.3} = YBa_{4}$ .

"Hole concentration per formula unit.

boundary effect. In the case of YBa\*, both the intrinsic quality and the grain boundary origin were mentioned (Yongz). The Hall coefficient  $R_{\rm H}$  decreases with decreasing oxygen content in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, with a plateau of very little change from  $\delta=0.1$  to  $\delta=0.4$  (Ongz1, Wangz).

Several papers (Allen, Alle1) have discussed the transport coefficients in the relaxation time approximation. They concluded that the Hall tensor tends to be holelike in the ab plane and electronlike in the ac and bc planes, which explains why sign differences have been reported in the literature.

#### 3. Thermoelectric Effects

A conductor with a temperature gradient and no electric current develops a steady-state electrostatic potential difference between the high and low temperature regions; this phenomenon is called the thermoelectric, thermopower, or Seebeck effect. This and other related effects such as the Peltier effect vanish in the superconducting state (Hundl).

Figure X-6 shows the temperature dependence of the thermoelectric power or Seebeck coefficient S of La<sub>2</sub>CuO<sub>4</sub> (Gran1) and (La<sub>1-x</sub>Ba<sub>x</sub>)<sub>2</sub>CuO<sub>4</sub> for several values of x (Alle3, Hundi, John3, Marcu, Uchi1, Uher1, Uher2, Yanz1). The coefficient decreases with increasing x=0.025, 0.05, and 0.075, and the latter two compounds exhibit a rapid decrease to zero near the phase transition. No drop was observed in S at low temperatures for x=0. In another study the same result was found for x=0.075, but S for the x=0 sample decreased dramatically to a very low value below 90 K. This was attributed to a changeover from an activated type of semiconductor transport at high temperatures to a variable range-hopping type at low temperatures (Maeno).

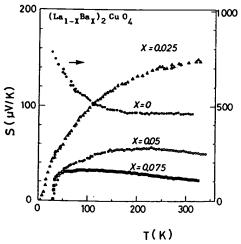


Fig. X-6. Temperature dependence of the thermopower S of  $(La_{1-x}Ba_x)_2CuO_4$  for several values of x (Uchi1).

A Hachment B page 13

The thermopower of polycrystalline granular  $(La_{1-x}M_x)_2CuO_{4-\delta}$  with M=Ba or Sr was calculated in the effective medium approximation and the results for the x dependence of S(T) are in good agreement with experimental values (Xiazz). The room-temperature thermopower data of  $(La_{1-x}M_x)_2CuO_4$  fit the Heikes expression (Coope, Heike)

$$S = -\frac{k}{e} \left[ 2 \ln 2 + \ln \left( \frac{2x}{1 - 2x} \right) \right] \tag{X-8}$$

where 1 - 2x is the number of electrons per copper site. Note that this expression has no adjustable parameters.

There are reports that above  $T_c$  the thermopower in YBaCuO and its rare-earth-substituted analogues is positive (Mawds, Mitra), and also is negative (Khimz, Yaozh, Yuzzz), and examples of both cases are shown in Fig. X-7. An electronic or negative S(T) was observed in  $O_2$ -heated superconducting samples, and holelike behavior with positive S(T) was seen in air-heated nonsuperconducting ones (Raych). A sample of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.9</sub> exhibited a negative thermopower between 300 and 125 K attributed to a diffusion mechanism, a positive S(T) from 125 to 90 K ascribed to phonon drag effects, with S=0 below  $T_c=90$  K (Yaozh; see also Khimz).

A large peak in the thermopower of YBa\*, called a precursor effect, was observed just above  $T_c$  (Mawds, Uher2). It was suggested that high- $T_c$  materials be used for thermopower test leads for the absolute determination of S(T) above the temperature range covered by the present standard Nb-Ti leads (Uherz).

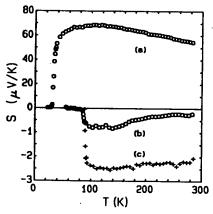


Fig. X-7. Temperature dependence of the thermopower S of (a) a ceramic sample of La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4- $\delta$ </sub>, (b) a ceramic sample of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, and (c) the ab plane of a YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single-crystal sample. Note the different scales on the positive and negative parts of the y axis (Yuzz3).

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#### 5. Thermal Conc

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Fig. X-8. Log-log inset shows the da

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### 4. Photoconductivity

Photoconductivity studies of LaCuO, YCuO, and YBaCuO samples suggest that polarons and excitons play a substantial role in the mechanisms of superconductivity (Masum, Masu1, Robas).

#### 5. Thermal Conductivity

The thermal conductivity K(T) is helpful in determining the fraction of the thermal energy that is transported by charge carriers and the amount carried by the lattice (phonons). It can provide information about the electron-phonon interaction, mean-free path, carrier density, and other physical properties. The thermal conductivity is not necessarily divergent or zero in the superconducting state. Thermal conductivity measurements have been reported on both polycrystalline and single-crystal samples (e.g., Bayot, More6, Uher2), and an example of the temperature dependence of K(T) in polycrystalline YBa\* is shown in Fig. X-8.

A detailed study of the transport properties of YBa\* (Gottw) included an analysis of the low-temperature (0.1 < T < 2 K) behavior of K(T) in terms of a phonon or lattice contribution  $K_{\rm ph}$  and an electronic contribution  $K_{\rm el}$ 

$$K(T) = K_{\rm ph} + K_{\rm el} \tag{X-9}$$

where

$$K_{\rm nh} = aT, \qquad K_{\rm el} = bT^3 \tag{X-10}$$

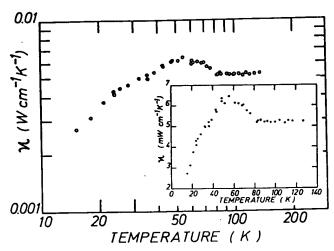


Fig. X-8. Log-log plot of the thermal conductivity of YBa\* versus the temperature. The inset shows the data replotted on a linear scale (More6).

A Hachmen + B page 15

with  $a=16~\mu W/K^2$  cm and  $b=47~\mu W/K^4$  cm. Hence even at  $\approx 0.1~K$  large portions of the thermal carriers appear to be in the normal state. An observed increase in K(T) below  $T_c$  indicated large electron-phonon scattering. The authors estimated  $v_F \approx 10^7$  cm/sec,  $N_{\rm eff} \approx 10^{22}/{\rm cm}^3$ , a small carrier mean-free-path, and pointed out the similarity with the heavy Fermion system CeCu<sub>2</sub>S<sub>4</sub>.

There was a report that  $K_{\rm el} \ll K_{\rm ph}$  at 300 K and  $n \approx 0.13$  carriers/Cu atom (Morió). The observed increase in K(T) below  $T_{\rm c}$  may be due to freer phonon flow, and suggests that a strong electron-phonon coupling is present in YBa\*. A  $T^3$  dependence of K(T) in YBa\* below 5 K provided evidence for a temperature-independent lifetime (Herem). At 50 K the Lorentz number  $L = K\rho/T$  was estimated to be  $26 \times 10^{-8}$  W $\Omega/K^2$ , a factor of 20 larger than the Wiedermann-Franz value, and  $K_{\rm el} \approx 0.1$   $K_{\rm ph}$  in polycrystalline (La<sub>0.9</sub>Sr<sub>0.1</sub>)<sub>2</sub>CuO<sub>4</sub> (Bartk). The thermal conductivity of sintered YBa\* in the temperature range from 0.1 to 10 K was linear in T at the lowest temperatures, and had a  $T^3$  dependence at the highest temperatures, in accordance with Eq. (X-9) (Graeb). This was claimed to be consistent with strong Rayleigh scattering of the phonons from the granular particles. In the ab plane single crystals of microtwinned HoBa\* obeyed a power law  $K(T) = aT^n$  with  $n \approx 2$ , a behavior similar to that observed in glasses (Graeb).

# D. TUNNELING PROPERTIES

Tunneling can be carried out through an insulating layer, I, between two superconductors (S-I-S), between a superconductor and a normal material (S-I-N), and between two normal materials (N-I-N) such as two semiconductors. The dc and ac Josephson effects involve particular types of tunneling phenomena across a barrier between two superconductors. The SQUID is an application of Josephson tunneling that involves macroscopic quantum phenomena. These topics will be discussed in the following three sections.

One of the preferred ways to measure an energy gap is through tunneling experiments, and many values of the gap energy determined by this technique are recorded in Table IX-1.

# 1. Tunneling Measurements

A tunneling study of LaSrCuO thin films was made using both the sandwich-type and the point-contact-type tunneling techniques (Naito). Sandwich-type junctions were prepared by forming a small window  $\approx 150~\mu m$  square and depositing a Pb counter electrode, which formed a natural high-resistance tunnel barrier, typically 1 M $\Omega$ . Point contact tunneling was done with the aid of a scanning electron microscope (SEM) using a tungsten probe with a tip radius less than 100 nm. Continuous scanning was not possible, but several points on the same specimen within a 1  $\times$  1  $\mu m^2$  window could be sampled. The curves of differential tunneling conductances dI/dV as a function of the bias voltage presented in Fig.

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Fig. X-9. Point at 4.2 K (Nait

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In a low-temperature SEM tunneling study of  $(La_{0.9}Sr_{0.1})_2CuO_{4-\delta}$  at 5 K the electrons tunneled from the tip into many superconducting grains with inhomogeneities small compared to the grain sizes (Kirtl). The data were analyzed in terms of the model of Zeller and Giaever (Zelle), and gave  $E_g/kT_c$  in the range from 3.5 to 6.3. Another experiment (Tsue1) was consistent with tunneling into grain sizes  $\leq 1$  nm which are much smaller than the apparent crystal sizes. A study of  $(Y_{0.55}Ba_{0.45})_2CuO_{4-\delta}$  using point-contact tunneling at 4.2 and 27.4 K provided gap ratios  $E_g/kT_c$  in the range 3.8-4.2, somewhat larger than the BCS ratio (Ekino). Figure X-10 shows an example of the way *I* versus *V* SEM electron tunneling curves give a range of gap values (5 meV  $\leq E_g \leq$  190 meV) for a niobium tip located at various points of an aluminum-doped YBa\* sample (Gall1).

Electron tunneling between the grains of oxygen-poor YBa\* exhibited a differential resistance dV/dI which appears to have an asymptotic peak that did not appear in homogeneous oxygenated samples (Escud). The gap signature was

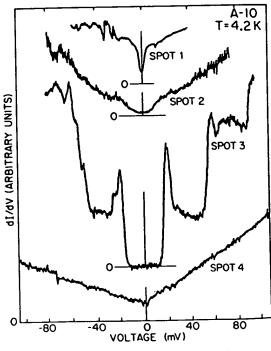


Fig. X-9. Point-contact tunneling conductance for four locations on a LaSrCuO thin film at 4.2 K (Naito).

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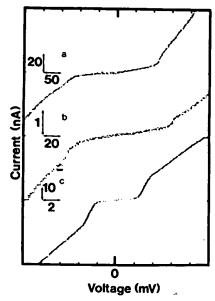


Fig. X-10. Current versus voltage plots obtained using a niobium tip at different positions on the surface of aluminum-doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub>. The difference scales used for each curve are indicated. The spectra from top to bottom show jumps in current at 95, 30, and 2.5 meV, respectively (Gall1).

not very well resolved and  $E_{\rm g}/kT_{\rm c}\approx 7\text{-}13$ . Tunneling studies of YBa\* thin films (Ogale) exhibited a dependence of the normalized critical current  $I_{\rm c}/I_{\rm c0}$  on  $T/T_{\rm c}$ , which differs from some recent S-I-S junction calculations (Ambeg). The deGennes model was reported to be better over a small temperature range, but the predicted curvature was larger than that of the data.

In the break-junction tunneling technique (More3, More5) a small piece of bulk material is electromechanically broken under liquid helium, and the freshly fractured surfaces are adjusted to form a tunneling barrier with the liquid helium acting as the insulator (More4). The most common current-voltage curves obtained by this technique (More4) closely followed a quadratic current dependence on the voltage. Some curves have derivatives that exhibit structure indicative of the presence of gap sum voltages, and others have shapes that suggest quasi-particle tunneling between superconducting electrodes (Morel). Related behaviors are observed, for example, for tunneling from a PtIr tip into LaSrCuO (Tsuei, Zelle).

In the absence of high-quality "sandwich" junctions these break-junction results are interesting. However, the effects of the strain and elastic failure on the electronic properties of the specimens and junctions is not clear. This could be of concern, particularly in the case of ceramic superconductors that are sensitive to sample treatment.

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#### 2. Josephson Effect

When two superconductors are separated by a thin layer of insulating material, electron pairs can tunnel through the insulator from one superconductor to the other. There are three effects of pair tunneling, namely:

- 1. The dc Josephson effect, which is the flow of a dc current  $J=J_0 \sin \delta$  across the junction in the absence of an applied electric or magnetic field, where  $\delta$  is a phase factor and  $J_0$  is the maximum zero voltage current.
- 2. The ac Josephson effect relates to the flow of a sinusoidal current  $J = J_0 \sin[\delta (4\pi eVt/h)]$  across a junction with an applied voltage V where  $\nu = 2eV/h$  is the frequency of oscillation.
- 3. Macroscopic quantum interference effects involving a tunneling current J with an oscillatory dependence on the applied field, given by

$$J = J_0 \frac{\sin \pi \Phi / \Phi_0}{\pi \Phi / \Phi_0} \tag{X-11}$$

where the magnetic flux  $\Phi$  may be approximated as the product of the average magnetic field strength times the cross-sectional area, and  $\Phi_0 = hc/2e$  is the fluxoid or quantum of magnetic flux.

In the reverse ac Josephson experiment, dc voltages are induced across an unbiased junction by introducing an rf current into the junction, or by radiatively coupling an rf signal through a coil surrounding the sample (Chenz, Weng1). It was suggested that this result supports the existence of granular superconductivity in YBaCuO at 240 K (Weng1).

Anomalous voltage excursions as a function of temperature and magnetic field strength were reported above 100 K (Caizz). The onset of the excursions was 20 mT, they reached a maximum at about 33 mT, and disappeared for fields above 56 mT. The voltage jumps were different for cooling and heating, and they were more frequent for larger samples. These transient voltages were attributed to flux jumps of granular superconductors. It was proposed that some grains have transition temperatures as high as 160 K.

Superconducting oxide materials are porous with chains of grains measuring a few microns in size. Owing to the inverse Josephson effect an applied rf current could cause individual Josephson junctions in these materials to develop quantum voltages given by  $V_j = nh\nu/2e$ , which is on the order of nanovolts. Such junctions could even be inside the grains themselves (Blaze). Thermal smearing can prevent the detection of individual quantum voltages, but observable dc voltages in the millivolt range can result from the summation of thousands of junctions with n values as large as 100.

Josephson junctions in thin films of LaSr(0.1) are believed to form at grain boundaries. In one experiment the Josephson current was found to be propor-

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c-junction failure on This could are sensitional to  $[1-(T/T_c)]^2$ . In contrast, samples of YBa\* (Cuizz) and BaPb<sub>1-x</sub>Bi<sub>x</sub>O<sub>3</sub> (Suzu2) produced current proportional to  $[1-(T/T_c)]^{3/2}$ , which may suggest proximity effects (Fuku3, Kobes, Kres3, Lynto) with comparatively long-range leakage of Cooper pairs (Moriw). The proximity effect can cause two superconductors with different  $T_c$  values in contact with each other to exhibit one intermediate  $T_c$ , and it can cause a nonsuperconductor-superconductor pair to act like a superconductor with a lower  $T_c$  (Kres3).

A LaSr\* sample mounted in a point contact current-voltage probe with a conducting tip produced a hysteretic I versus V characteristic of the type shown in Fig. X-11 (Estev, Tsaiz). Microwave irradiation produced the Shapiro steps (Baron), which result from the beating of the oscillating Josephson supercurrent with the microwaves. The separation in voltage between these steps is proportional to the microwave frequency, and their amplitude is Bessel-like. The Josephson junction characteristics are observed even when the metal tip is nonsuperconducting, which indicates that the superconducting junction is inside the material under the tip (Estev). The absence of voltage steps during the microwave irradiation of a Nb-YBaCuO point-contact junction suggests that the coupling between the superconducting regions along the percolative path is non-Josephson (Tsaiz). Other workers studying Nb-YBa\* point-contact junctions observed Shapiro steps and an unusual noise behavior along the I-V characteristic (Kuzni), and an estimate was made of  $\Delta \approx 19.5 \pm 20$  mV and  $2\Delta/kT_c \approx 4.8$ (Baro1). Very clear steps were observed in a weak link fabricated by carving a bridge (0.1 × 0.2 mm<sup>2</sup>) in a 10 × 2 × 2 mm<sup>3</sup> YBa\* sample. Both harmonic  $(n\Phi_0)$  and subharmonic  $(n\Phi_0/m)$  flux quantum steps were reported at 77 K (Chan2). Other microwave radiation experiments have also been carried out (McGra, Nieme, Mengz).

Theoretically microwave absorption by S-I-S junctions with square-well potentials at low temperatures is calculated to occur in sharp steps whenever the microwave energy is an integral multiple of the minimum energy needed to excite quasi particles from the ground state to bound Andreev excited states (Aberl, Andre). Andreev reflections at an Ag-YBa\* interface on a thin film have been reported. It was argued to provide evidence for Cooper or zero-momentum carrier pairs. The gap as determined from the reflection data was 12.5 meV, compared with 14 meV deduced from tunneling on the same film. These results are consistent with a BCS picture (Hoeve).

# 3. Macroscopic Quantum Phenomena

Macroscopic quantum phenomena were reported in Sn-YBaCuO and YBa-CuO-YBaCuO point contacts. The critical current and the voltage are periodic in the magnetic field, and for each voltage there is a minimum and a maximum value of the current between which the oscillations take place when the magnetic field is varied (DeWae, DeWa1). This dc SQUID (Finkz, Jakle, Soule) behavior has been observed up to 40 K (Kawab), 66 K (DeWae, DeWa1), and 90 K (Tsai1).

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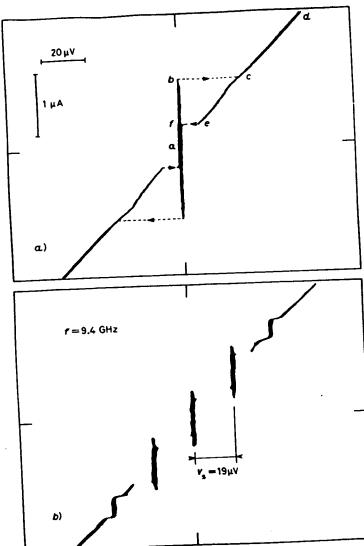


Fig. X-11. (a) Oscilloscope trace of a current-voltage characteristic obtained at 4.2 K with an aluminum tip on a LaSr\* sample. Letters a through f indicate the sense of the trace. Dashed lines have been added to indicate the switching between the two branches. (b) Steps induced by microwave irradiation at the frequency f = 9.4 GHz (Estev).

SQUIDs have been fabricated from YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Colcl, Kawab, Zimme). The flux quantum  $\Phi_0$  obtained from these devices was the expected hc/2e (Gough, Koch1, Koch2). For a YBa\* specimen to exhibit SQUID behavior the sample need not necessarily be multiply connected. For such a YBa\* SQUID operating in the magnetometer mode the field spectral density was  $5.8 \times 10^{-10}$   $T/\sqrt{H_C}$  at 4.2 K and  $1.5 \times 10^{-10}$   $T/\sqrt{H_C}$  at 77 K (Pegru). This value is better than that of a good flux-gate magnetometer.

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# ATTACHMENT C

# NATIONAL SCIENCE FOUNDATION WASHINGTON, D.C. 20550

Dem Drs. Bednoz + Miller. 12/3/86 This is just to suform you that my group at the U. of Howton has seproduced your Nesults (2. Thys. B 64, 189 (&)) Three weeks ago. A small ac diamagnetic signal was also detected. Magnetic field was found to suppress The transation. I believe that it is superconducty. Now The question is "what place" or "mused places: Soon, you will here from use more. Please send me more information! Sincerely yours my phone: (202) 357-9737 C.W. Chu

P.S. Currently, I am the Director of Solid Physic Program

(also Physics. Univer Housan)
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